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A Literature Review on Radioactivity Transfer to Plants and Soil

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A LITERATURE REVIEW ON RADIOACTIVITY TRANSFER TO PLANTS AND SOIL

Ole John Nielsen

Abstract. The literature on transfer of radioactivity to plants and soil have been reviewed. Main emphasis has been placed on the transfer to plants. A general plant transfer model has been set up. Root uptake, dry deposition, wet deposition, weathering, resuspension, and translocation are the transfer processes discussed. Theory and experimental data for evaluating the different pathways are reported. Only iodine-131, strontium-90 and caecium-137 have been considered in the report. Recommendations of specific transfer parameters have been given as realistic as possible. Suggestions for further research have also been given.

INIS descriptors: CESIUM 137; DEPOSITION; IODINE 131; MATHEMATICAL MODELS; PARTICLE RESUSPENSION; PLANTS; PRECIPITATION SCAVENGING; RADIONUCLIDE MIGRATION; RETENSION; REVIEWS; ROOT ABSORPTION; SOILS; STRONTIUM 90; TRANSLOCATION; WASHOUT

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1. PREFACE

The report reviews literature on radioactivity transfer to plants and soil; the main emphasis has been placed on the transfer to plants. The report contains a survey of the different transfer pathways. Theory and experimental data for evaluating the different pathways are reported. Recommendations for parameter values for use in risk assesments are given.

As an aid in identifying the literature two literature searches were performed: one on the INIS data base in Vienna and the other on the PASCAL data base in Paris. Personal contact has been made with several European laboratories as well as with institutions outside Europe. A considerable effort is currently being concentrated on this subject, and new results continue to arrive as this report is being completed.

This review gives 180 references. Of these approximately half are European and half are from outside Europe.

2. INTRODUCTION

Evalutation of population hazards following a hypothetical nuclear power plant accident is a very complex task. Some of the radioactive effluents, discharged to the atmosphere, will be transferred through the biosphere to man, who can be exposed by ingestion of contaminated terrestial foodstuffs. Even though the intermediate transfer of radioactivity to plants is only a small part of the total consequence model, it is very complex in itself.

In this study only short-term effects are studied This means that only time intervals up to one year are considered even though dealing with the long-lived nuclei, ^{90}Sr and ^{137}Cs. Before proceeding to describe the plant transfer model some of the assumptions in the plant transfer model should be mentioned.

A plant transfer model has been set up (see figure 3.2 in chapter 3). For the sake of completeness all transfer processes, irrespective of their relative importance, have been shown in the figure. The number of a transfer process (TP) in figure 3.2 corresponds to the number of the chapter dealing with the respective transfer process.

The plant transfer model makes three assumptions. First, the concentration, $X_i(x,y,z,t)$, of material i in the plume is known as a function of position and time as are the chemical and physical form of the radionuclides. Second, the vegetation type, density and distribution are known. Third, the weather - wind speed, humidity and precipitation type, rate and spectrum (e.g. raindrop size distribution) - is known as a function of position and time.

Only a few radionuclides contribute significantly to internal dose commitments. This is due to many different factors, among others the radioactive halflife, the physico-chemical form of the radionuclides, the release fraction, the metabolic behaviour in plants, animals and man. Knowledge of the importance of the different radionuclides is available from experimental studies on nuclear weapons fallout in man's diet.

In this study only iodine-131, strontium-90, caesium-137 and plutonium are considered. Iodine is specificly absorbed in the thyroid gland. Strontium is specifically transferred to the bone. Caesium is absorbed in muscle tissue and causes doses to the whole body.

3. PRESENTATION OF THE PLANT TRANSFER MODEL

Like most other phenomena occurring in nature, transfer of radioactivity to plants is described in terms of models. Before giving the description of a plant transfer model some general aspects of models will be mentioned.

In the most simple sense a model can be described by a mathematical formulation which allows an quantity, or dependent variable, to be expressed in terms of known or measurable variables. This is illustrated in figure 3.1, where the line symbolizes a mathematical expression. As most other models the plant transfer model is a composite model consisting of a number of submodels.

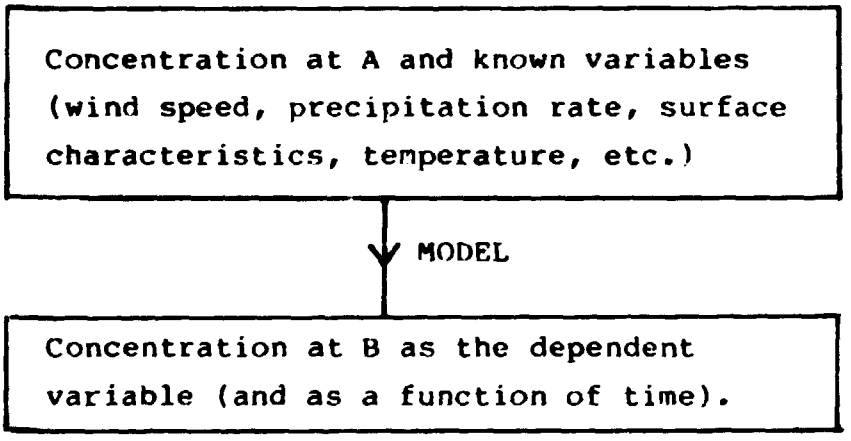

Figure 3.1. Schematic illustration of a model.

The plant transfer model is illustrated schematically in figure 3.2. For the sake of completeness all transfer processes, no matter how important they are in this study, are considered in the diagram. The number of the transfer process (TP) in the diagram corresponds to the number of the chapter concerning the process.

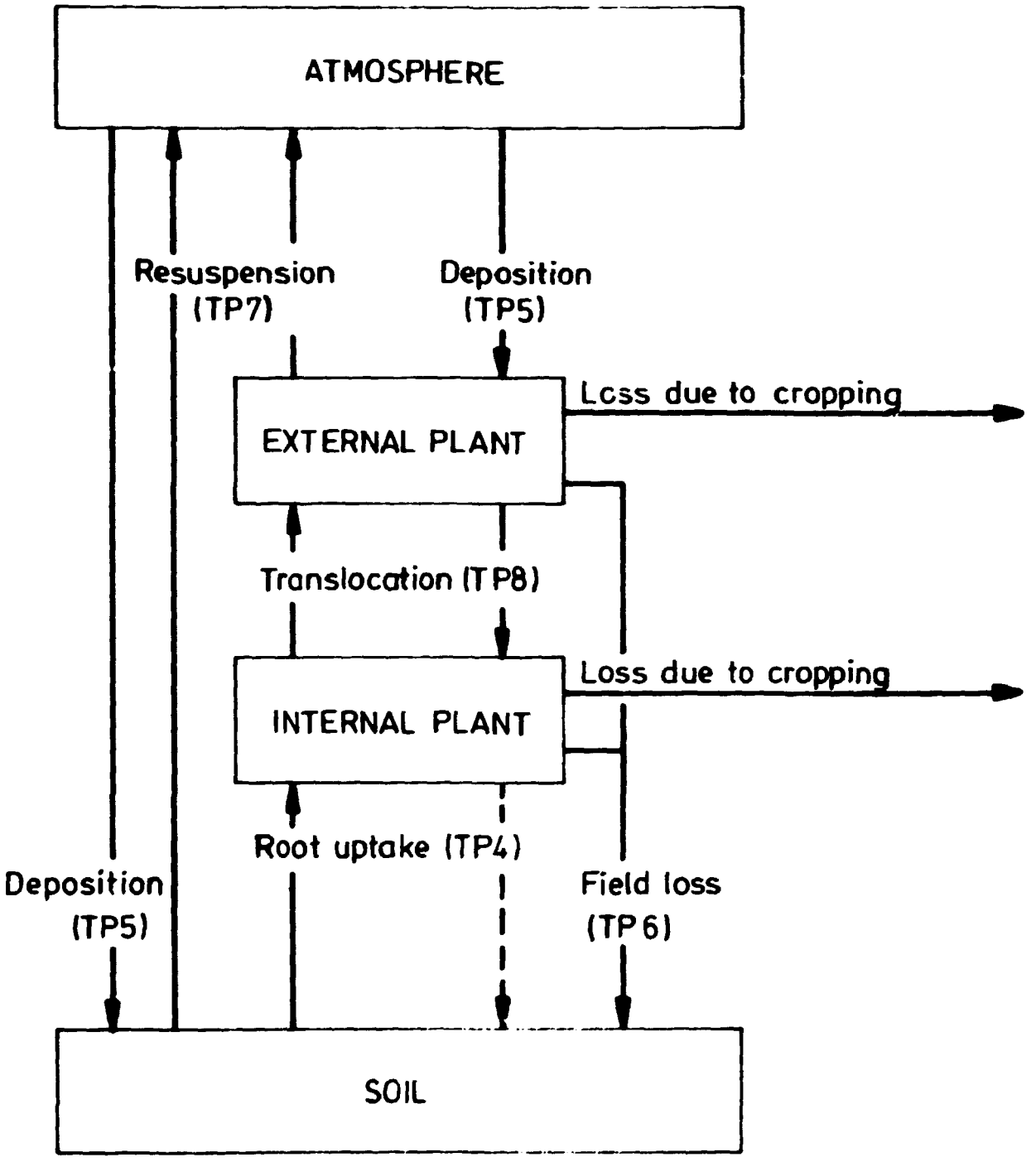

Figure 3.2. Schematic representation of the plant transfer model. Numbering corresponding to the number of the chapter dealing with the respective transfer process.

Radioactive effluents discharged to the atmosphere may be deposited on the soil and on the external parts of the plants (TP5). Deposition may occur in the absence or presence of precipitation, (dry or wet deposition). Loss of activity (TP6) from the exterior of the plant is divided into two parts depending on time. A certain part of the deposited material is initially lost or as it is more commonly expressed, there is an initial fractional retention of material on the vegetation. Some part of the initially retained activity is removed slowly by various weathering processes. Radioactive material may be translocated (TP8) to the interior of the plant. Soluble material may be transported from the soil to the interior of the plant by root uptake (TP4). Activity is of course removed when plants are harvested or eaten by grazing animals, cropping. Cropping can also cause loss of activity from the external parts of neighbouring plants, which are not cropped. Dilution due to growth is not a transfer process, but the concept is important when considering activity per gram plant matter.

No evidence of transfer from the interior of plants to the exterior and to the soil has been found in the literature reviewed. All transfer pathways other than the two just mentioned are considered in this study. Plant decay may be considered as transfer from the interior of plants to soil. This pathway is not considered in the study. Theoretical and experimental data showing the importance of the different pathways are reported.

4. ROOT UPTAKE

4.1. Introduction

When *short-term* health hazards after a release of radioactive effluents are evaluated, transfer of radioactivity from soil to plants via root uptake is normally neglected.

If radionuclides are to be absorbed through the roots they must be deposited in a soluble form or they must be made soluble in the soil. Factors affecting the root uptake of radionuclides from soil by plants have been the subject of a number of reviews (Men65, Rus66). Absorption of elements from soil by plants depends on many factors: soil type, climate conditions, plant type, chemical form of radioactivity, time of deposition relative to plant growth cycle and distribution of activity within the soil.

With the assumption of a few percent uptake during one growing season, it has been calculated (Was75) that residence in an area, with enough soil contamination to produce unacceptable doses by root uptake and ingestion of food plants, would be restricted already by other restrictions.

Because of the slow rate of root uptake a short lived nuclide like I-131 (half-life = 8 d.) need not be taken into account. Only caesium-137 (half-life = 30 y.) and strontium-90 (half-life = 28 y.) will be considered further. Strontium is not like caesium strongly adsorbed to clay mineral soils. Caesium is not strongly absorbed by organic soils (i.e. soils with high content of organic matter). Strontium has the potential of contaminating foodstuffs via root uptake and concentrates in bone. These are the reasons that most experimental work is done on strontium. Research on factors that affect the availability of strontium to root uptake by plants has recently been critically reviewed by Francis (Fra78).

Lassey (Las79) has treated the general problem of predicting dietary contamination from root uptake. He concludes root uptake to be without importance for at least the first months after deposition.

4.2. Review of experimental data

Only a few representative experimental data on strontium uptake will be presented here. Andersen (And67) presents a vast amount of data on the uptake of strontium and caesium by 44 different plant species and 22 different Danish soil types. Some results show that the radionuclide concentration per gram dry matter of plant varies by orders of magnitude with time of year within 600 days after deposition. This cannot be explained only by dilution because of plant growth. Maximum root uptake of strontium during one growing season is approximately 7% of the soil content, observed for red clover. Romney et al. (Rom63) found that 0.10% to 0.15% of the beta activity (Sr-89, Sr-90, Ce-144, Cs-137, Ru-106, Y-91) added in soluble form to the soil surface was removed by wheat grown for 117 days. Milbourn (Mib60) reports 2% uptake of Sr-90 by rye grass during the first year after deposition. Van der Stricht et al. (Van71) estimated a maximum annual grass root uptake of 5% for Sr-90, analysing several years of fallout data.

4.3. Conclusions

Uptake of nutrients by roots and flow of nutrients upwards in plants vary with time of the year. Hence, time of deposition is likely to have an appreciable effect on root uptake rate, especially during the first year. However, no experimental data illustrates this in detail and this is normally not taken into account by prediction models.

It can be concluded that root uptake will be an unimportant transfer pathway as compared to direct contamination when evaluating health hazards during the first year after deposition.

5. DEPOSITION ON PLANTS

Matter, particulate or gaseous, is transferred from the atmosphere to plants by a number of different processes. These processes are collectively called deposition. All these different processes depend on three types of parameters: parameters describing the matter to be deposited, e.g., physico-chemical parameters (particle size distribution etc.), meteorological parameters, e.g., temperature, wind velocity, humidity etc. and surface parameters, e.g., vegetation type, density, physiology etc. Transport of matter to plants is not only studied with special reference to radioactive effluents, but also with reference to other kinds of air pollution. The term deposition is very often used as a convenient way to characterise the entire complex of physical phenomena that may result in transfer of matter to the ground at the air-ground interface including the absorption of gases. The very important concept of deposition velocity V_g was introduced by Chamberlain in 1953 (Chm53a). He defined V_g either as

$$V_g(z) = \frac{F}{X(x,y,z)} ,$$

where F is the flux to the surface, defined as the amount (g or curie) deposited per unit area per unit time, and $X(x,y,z)$ is the volumetric concentration at some reference point (x,y,z) above the surface, or as

$$V_g(z) = \frac{F_T}{X_T(x,y,z)} ,$$

where F_T is the total deposition per unit area (g or curie) and $X_T(x,y,z)$ is the time integrated concentration (TIC). V_g is a

function of reference height. This height dependence is not of practical significance in numerical codes.

Experimental values of deposition velocities can be obtained from different kinds of experiments. Field experiments involve releases of matter and measuring deposition and concentration. Values of V_g have also been obtained in connection with two reactor accidents, Windscale, 1957, and the SL1 reactor at NRTS, Idaho, USA 1961. Wind tunnel and other types of laboratory experiments are also sources of V_g values. One of the major difficulties in some field experiments is the uncertainty on the chemical and physical form of the material deposited. This is specially the case for iodine.

V_g has been shown to be highly dependent upon the density, height, growth form and areal distribution of vegetation (see, e.g., Ada71, Bun 66, Bun 68, Chm70, Haw64, Hei74, Zim69). Experimental deposition data are therefore sometimes given in terms of the normalized deposition velocity, V_D, defined as V_g (for vegetation) normalized to vegetation density. In practice it is determined through the relationship

$$V_D \ (cm^3 g^{-1} s^{-1}) = C_V \ (Cig^{-1}) : X_T \ (Ciscm^{-3}) \ ,$$

where C_V is activity per unit mass vegetation. In a series of field tests with stable elemental iodine, V_D ($cm^3 s^{-1} g^{-1}$ dry weight) was found to be significantly less variable than V_g (cms^{-1}) for pasture grass and clover cut one inch above the ground (Vog73, Hei74). It may be preferable (Hof75) to measure deposition per unit dry weight rather than per unit wet weight, because the moisture content of herbage is highly dependent upon air temperature, time of the year, air and soil humidity. The advantage of using V_D instead of V_g is that the need to estimate an effective area cropped by humans or animals is avoided. Angeletti (Ang80) argues strongly against using dry weight. He has shown that V_D values ($cm^3 s^{-1} g^{-1}$ dry weight) measured in summer vary distinctly from measurements in the rest of the year. He comments that since usually fresh grass or vegetables are consumed, deposition should be measured per unit wet weight.

To get a more detailed knowledge of the deposition phenomenon it is appropriate to divide the deposition processes into four groups: dry deposition of gases, dry deposition of particles, wet deposition of gases and wet deposition of particles. It has been found that 11% of the total particulate deposit has been deposited in absence of precipitation - dry deposition (Aar79). These processes are now considered in turn.

5.1. Dry deposition

5.1.1. Dry deposition of gases and vapours

5.1.1.1. Introduction. When the concentration of the gas to be deposited is not too high, the rate of deposition on the surface is assumed to be proportional to the concentration adjacent to the surface. The flux can be written as already mentioned

$$F = X(x,y,z) \cdot V_g(z) \quad . \tag{1}$$

The turbulence in the air depends on the distance from the surface. In order to derive formulations of deposition velocity, it is necessary to organize the atmosphere-surface system into layers where different transport processes, described empirically or theoretically, dominate. Each region can be said to have a resistance to transport, analogous to electrical resistance. Most analyses of transport to the surface assume that the surface and the air properties are homogeneous within each layer. Because of these horizontal layers the different resistances can be regarded as being in series. The flux can also be expressed in terms of a total resistance, r(z)

$$F = X(x,y,z)/r(z) \tag{2}$$

$$V_g(z) = 1/r(z) \tag{3}$$

where

$$r(z) = r_a(z) + r_b(z) + r_c(z) + \ldots\ldots , \qquad (4)$$

$r_i(z)$ being the resistance of horizontal layer "i". The advantage of expressing the transport in terms of resistance rather than in terms of velocity is that the resistances of different regions may be considered to be additive.

The total resistance in a complex atmosphere-surface system can of course not be predicted theoretically and will later be discussed on the basis of experimental results. However, it is possible to make some predictions from the framework of micrometeorology, provided the condition of horizontal homogeneity is satisfied.

The first region to be considered is the atmospheric turbulent surface layer, where eddy diffusion dominates. In this region molecular properties play no part and transport is expected to be the same for all gases and vapours and similar for heat and momentum. In this layer the vertical gradient of the wind speed u is given by

$$\frac{\partial u}{\partial z} = \frac{u_*}{k \cdot z} \phi_M \left(\frac{z}{L}\right) , \qquad (5)$$

where k is the so-called von Kármán constant, k = 0.4, (Lum64). The friction velocity u_* (a turbulent velocity scale) is defined in terms of the surface drag τ due to the wind

$$\tau = \rho u_*^2 , \qquad (6)$$

where ρ is the density of air. ϕ_M is a correction for atmospheric diabatic effects, and L is the Monin-Obukhov length (Lum64), which depends on the heat flux in the surface layer. In the limit as $z/L \to 0$, ϕ_M is equal to 1 and (5) is easily integrated

$$u(z) = \frac{u_*}{k} \ln \frac{z}{z_o} \qquad (7)$$

The integration constant z_o is the height at which u vanishes. z_o is called the roughness length. For a real grass surface or canopy of height h, the wind profile will look like the one given in figure 5.1. The wind speed will not be logarithmic in z, but in z-d, where d is a constant normally called the zero plane displacement, and d can be determined experimentally. z_o then is the height above d where the extrapolated wind speed becomes zero. Since d is only a zero point displacement, d is omitted in this context.

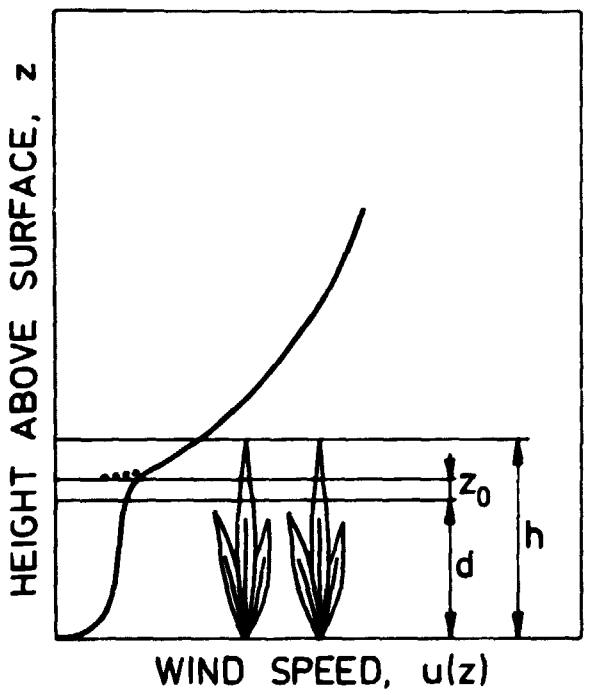

Fig. 5.1. The wind profile over a rough surface shown schematically.

Under diabatic conditions one can express the solution as a logarithmic term plus a diabatic correction

$$u(z) = \frac{u_*}{k} \ln \frac{z}{z_o} + \psi_M(\frac{z}{L}) \quad . \tag{8}$$

Similar to (5) the concentration gradient $\partial X/\partial z$ of a particular constituent is given by

$$\frac{\partial X}{\partial z} = \frac{X_*}{k \cdot z} \phi_C(\frac{z}{L}) \quad , \tag{9}$$

where X_* is a characteristic scale for turbulent concentration fluctuations such that $X_* u_*$ represents the vertical flux of the constituent. ϕ_C is also here a correction for diabatic effects. Integration of (9) gives

$$X(z)-X_o = \frac{X_*}{k} \ln \frac{z}{z_{oc}} + \psi_C(\frac{z}{L}) \quad . \tag{10}$$

$X(z)$ is seen to be a logarithmic function of height. X_o is the surface concentration ($z = z_{oc}$). z_{oc} is of similar magnitude as the roughness length but does not necessarily have exactly the same value. z_{oc} can be called the boundary surface transfer length. $\ln(z/z_{oc})$ can always be written as $\ln(z/z_o) + \ln(z_o/z_{oc})$. It is seen that the first term will dominate at sufficient height of observation, whereby any difference between z_o and z_{oc} becomes less significant.

The vertical flux $X_* u_*$ (see above) can also be expressed as $V_g(z) \cdot X(z)$, hence (Jen80a and Wes77b) $V_g(z)$ and $r(z)$ can be written

$$V_g(z) = \frac{ku_*}{\ln \frac{z}{z_o} + \ln \frac{z_o}{z_{oc}} + \psi_M + k \frac{X_o}{X_*}} \tag{11}$$

$$r(z) = (ku_*)^{-1} \ln \frac{z}{z_o} + \ln \frac{z_o}{z_{oc}} + \psi_M + k \frac{X_o}{X_*} \quad . \tag{12}$$

The term kX_o/X_* represents the fact that the surface cannot always absorb all material that is transported to the surface by turbulent diffusion. This term will be dealt with later in this chapter.

The resistance r(z) can be divided into a turbulent boundary resistance $r_a(z)$, a laminar sub-layer resistance $r_b(z)$ and a surface resistance $r_c(z)$, which is also called canopy or stomatal resistance

$$r_a(z) = (ku_*)^{-1} \left(\ln \frac{z}{z_o} + \psi_M\right) , \tag{13}$$

$$r_b(z) = (ku_*)^{-1} \ln \frac{z_o}{z_{oc}} , \tag{14}$$

$$r_c(z) = (ku_*)^{-1} k \frac{X_o}{X_*} . \tag{15}$$

Let us consider a case where there is no significant difference between z_o and z_{oc}, and where diabatic influence is ignored. In this case $V_g(z)$ has maximum for $X_o = 0$, which corresponds to a perfect absorbing surface. A perfect absorber can, for example, be realised in the case of a gas being highly soluble in or highly reactive towards the surface medium. It is very conservative to assume $X_o = 0$. However, this leads to

$$V_g(z) < u_* \frac{u_*}{u} , \tag{16}$$

$$r_a(z) > u_* \frac{u_*}{u} . \tag{17}$$

For typical values (z_o = 1 cm, u = 5 m/s, z = 10 m) we have u_* = 0.3 m/s. V_g would then be 1.8 cm/s at the most. In practice, with $X_o \neq 0$ the deposition velocity would be a lot less. The effect of ψ is to further reduce V_g during stable conditions. For unstable conditions the trend would be the opposite.

The laminar sub-layer resistance to transfer through the layer of air in contact with the surface, $r_b(z)$ can be expressed in terms of a surface transfer function defined by

$$r_b k u_* = \ln \frac{z_o}{z_{oc}} = k \cdot B^{-1} \quad , \tag{18}$$

where B is the dimensionless sub-layer Stanton number (Owe63). In most formulations, B^{-1} is written as proportional to fractional powers of both the Schmidt number, Sc = ν/D, and the surface roughness Reynolds number, Re = $u\, z_o/$,

$$B^{-1} \sim \frac{u_* z_o}{\nu}^{m} \frac{\nu}{D}^{n} \quad , \tag{19}$$

where ν is the kinematic viscosity of air, D is the relevant molecular diffusivity and m and n are constants. Experimental evidence suggests that B^{-1} varies little with wind speed and plant geometry (Chm75a). r_b values can be estimated from

$$r_b(z) = (B u_*)^{-1} \tag{20}$$

Experimental values of B^{-1} lie in the range from 2 to 40.

The residual resistance, r_c, to transport at or in the cuticle of the vegetation, is called surface resistance, r_c depends on such factors as the type, age and condition of the vegetation, water availability, solar radiation level, air temperature etc. Some of these factors control opening and closing of stomata (Amb66). This of course influences stomatal resistance of growing plants. It is extremely difficult to generalize about values of stomatal resistance except for very special situations. For a dense canopy with leaf area index (LAI, leaf area per unit soil area beneath) of at least 2.5, minimum values of r_c (giving maximum V_g values) for water vapour have been summarized by many authors, among them Holmgren et al. (Hol65), who report r_c values from 0.2 to 3.4 s cm^{-1} for various types of vegetation. Stomatal resistance is inversely proportional to molecular diffusivity. Values of D for various gases are given in table 1.

Table 1. Values of molecular diffusivities D in $cm^2\ s^{-1}$

Iodine	0.08
Water	0.25
ThB ions	0.054
CO_2	0.14
SO_2	0.12

The effects of having wet or waxy vegetation will be discussed later.

5.1.1.2. Review of experimental data. The most important gases and vapours to be considered are elemental iodine and vapours of chemical compounds containing iodine e.g. methyl iodide, CH_3I. Noble gases are normally not considered to deposit even though xenon-133 has been detected on airfilters and aircrafts after nuclear test explosions (Aar67). One of the main difficulties in doing experiments on dry deposition of gases is knowing what fraction of the originally releases gas, iodine, remains in the gas phase at the site and time of deposition. This is due to the fact that iodine vapour absorbs on particles. The results of measurements of dry deposition of gaseous I_2 and CH_3I are summarized in table 2. Experimental details of the measurements must be sought in the original references.

The first experiment done using the concept of deposition velocity is reported by Chamberlain and Chadwick in 1953 (Chm53b). Compared with the rest of the V_g values in table 2, their measured values seem to be very high. There is no obvious reason for this. Deposition velocity to grass is reported to be 3-5 times that to filter paper. However, this does not mean that iodine has a greater affinity to grass than to filter paper, but the effective area of grass leaves corresponding to 1 cm^2 of soil area is much greater.

Table 2. Measurements of gas deposition velocities.

Reference	V_g (cm s^{-1})	Gas	Surface	Type of experiment	Comments
Chamberlain (1953)	1.72 - 3.75	131-I_2	grass	field	u: 1.4 - 4.4 m/s . u_* = 15 - 40 cm/s
	0.30 - 0.94		filter paper		z_o: 1.2 - 2.8 cm . V_D is per unit wet weight
Stewart (1958)	0.30 - 0.11	131-I_2	grass	field	Windscale accident
Chamberlain (1959)	0.36	131-I_2	grass	field	Windscale accident
Chamberlain (1960)	1.2	131-I_2	bean leaves	wind tunnel	
	2.1		paper leaves		
	0.3		leaves	field	u = 3.4 m/s . z_o = 1.0 . u_* = 26 cm/s
	0.6		paper leaves		
Barry (1963)	only relative and qualitative measurements	132-I_2	bean leaves	wind tunnel	
Hawley (1964)	0.6	131-I_2	grass	field	
Cline (1965)	0.55	131-I_2	grass (unclipped)	field	
	0.59		grass (clipped)		
	0.52		sagebrush leaves		
	0.35		dry grass		
	0.33		dry soil		
Chamberlain (1966)	results given in terms of B^{-1}	ThB	different	field, wind tunnel	
Hull (1966)	0.25	131-I_2	grass	field	from fall-out data from a Chinese weapons test.
Chamberlain (1966)	1.5	131-I_2	grass	field	
	0.54	131-I_2	grass	field	from fall-out data
	0.16	I_2	grass	field	analysis of stable iodine measurements
Bunch (1966)	0.82	131-I_2	grass	field	
Hawley (1966)	0.60 . 0.90	131-I_2	grass	field	
	0.84 . 0.38		soil		
	0.14 . 0.41		sticky paper		
	0.11		snow		
Atkins (1967)	(8.6 - 25) x 10^{-5}	CH_3I	different	wind tunnel	
	(1.4 - 24) x 10^{-4}		grass	field	
Bunch (1968)	0.1 - 3.5	131-I_2	grass	field	different meteorological conditions
	10^{-4}	CH_3I	grass	field	different meteorological conditions
	0.34	131-I_2	snow	field	
Zimbrick (1969)	1.2	131-I_2	grass	field	u_* = 60 cm/s. Treatment of many data in this paper.
Adams (1971)	results given in terms of a transfer parameter, P_t	131-I_2	grass	chamber	dependence in stomatal opening verified.
Vogt (1971)	0.04 - 0.58	131-I_2	grass	clovebox	
Vogt (1973)	results given in terms of V_D	CH_3I	grass	clovebox	
	0.20 - 1.54	131-I_2		field	
	1.29	131-I_2	clover	field	
Vogt (1974)	0.4 - 1.1	131-I_2	grass	field	this paper also treats the data from Vogt (1973)
	1.4 - 1.0		clover	field	
Heinemann (1976)	0.47 - 2.83	131-I_2	dry grass	field	this report also treats the data from Vogt (1973) and Vogt (1974)
	0.88 - 6.27	131-I_2	wet grass	field	
	4.22 - 2.20	131-I_2	dry clover	field	

In connection with the accident at Windscale (UK) on October 10, 1957 Stewart and Crooks (Ste58) obtained V_g = 0.30 cm/s near Preston 100 km south-east of Windscale and 0.11 cm/s in southern England. These values derived under meteorological conditions identical to those of Chamberlain and Chadwick are considerably smaller. Chamberlain measured a V_g value of 0.36 cm/s also in connection with the Windscale accident (Chm59). He also changed Stewart and Crooks' value of 0.30 cm/s to 0.44 cm/s from a difference in the method of working back the results from the date of measurement to the date of the accident. In the very extensive work from 1960 Chamberlain found V_g values of 1.2 and 2.1 cm/s for bean and paper leaves respectively in wind tunnel experiments (Chm60). In field experiments reported in the same paper the V_g values for the same materials were found to be 0.3 and 0.6 cm/s respectively. From autoradiographs of leaves it was found that iodine enters the leaves mainly at points where the skin is damaged. Also it was concluded that a waxy cutin on the leaves inhibits adsorption of iodine to some extent. From the wind tunnel experiments of Barry and Chamberlain (Bar63) it has been concluded that deposition of iodine vapour (filtered for particles) depends markedly on humidity. V_g to bean leaves in humid air was derived to be approximately 0.8 cm/s. In dry air it falls to less than one twentieth of that value, to V_g = 0.03 cm/s. This may indicate that uptake of iodine vapour is controlled by stomatal opening, which increases with humidity. Cline found deposition velocities around 0.6 cm/s for living grass and sagebrush and 0.4 cm/s for dry grass in field experiments (Cli65). A very extensive research program has been performed at The National Reactor Testing Station, Idaho, called the controlled environment radioiodine tests (CERT) project. It consisted of a series of planned releases of radioiodine over different vegetation and during various meteorological conditions, with the objective of estimating the transfer of radioiodine through the air-vegetation-milk-human chain. During the CERT project, 1963 to 1969, a lot of field deposition experiments have been performed and results reported (Haw64, Ada66, Bun66, Bun68, Zim69). In the first report (Haw64) deposition velocities to grass ranged from 0.4 to 0.8 cm/s with an average of 0.6 cm/s. In the second report (Ada66) The average

is 1.0 cm/s for the deposition velocity to grass in September, but for a higher average grass density. In the third report deposition velocities for CH_3I are reported to be at least a factor of 100 less than for iodine vapour. Experiments have been carried out for various grass densities up to 2000 g/m^2 wet weight, and V_g for a wind speed of 1.1 m/s has been related to grass density W_w (wet weight) by the equation

$$V_g = 6.3 \times 10^{-4} W_w^{0.837} \quad , \tag{21}$$

where V_g is in cm/s and W in g/m^2, and it should be noticed that the formula is not dimensionally correct.

It was found that deposition velocities based on dry weight were much less variable than those based on fresh weight. This could be expected since dry weight must be a more realistic indicator of plant area. For normal dry weight grass densities 100-200 g/m^2 one can use a proportionality with W_d^1 in place of $W_w^{0.837}$ given in (21). For more dense pastures (21) should be followed. Empirical analyses indicate that V_D values in $cm^3g^{-1}s^{-1}$ for W_w up to 2000 g/m^3 can be estimated from the following formulas when either u_* or u is known in m/s

$$V_D = 176\ u_* \quad , \tag{22}$$

$$V_D = 18\ u \quad . \tag{23}$$

These formulas are also not dimensionally correct and imply a given height and density.

In the fourth report (Bun68) V_g is fitted to dry weight grass density by

$$V_g = 0.0096\ W_d \tag{24}$$

in usual units. All dependences in this formula are put into the proportionality constant. Apart from this, earlier CERT data are summarised in this report. In the last report V_D was found to correlate with (u_*^2/u) in cm/s and wind speed u in m/s

$$V_D = 14.6 \cdot (u_*^2/u) \quad , \tag{25}$$

$$V_D = 18.5 \cdot u \quad . \tag{26}$$

V_D was also found to correlate with the boundary layer resistance model.

Hull (Hul66) determined an iodine deposition velocity of 0.25±0.10 cm/s from fallout data after a Chinese weapon test in October 1964. He also observed that the amount of particulate iodine varied significantly. From 70 per cent the first day of observation to around 30 per cent the following days. The deposition velocity is more a grand mean value than specific for gaseous deposition.

In a later paper of Chamberlain (Chm66a) deposition velocities vary by an order of magnitude: field experiments, V_g = 1 cm/s; fallout measurements, 0.5 cm/s; stable iodine measurements, 0.16 cm/s.

The results of the wind tunnel experiments performed by Chamberlain (Chm66b) with ThB vapour are given in terms of B^{-1}. B^{-1} showed little variation with values of u_* and z_o (u_* = 10-200 cm/s, z_o = 0.05-1.00 cm) applicable in normal conditions for vapour transport to grass in the field. B^{-1} was found equal to 8±1. Extension to field conditions is not straightforward and results in terms of B^{-1} not very useful (Pas74).

Atkins (Atk67) determined deposition velocities for CH_3I to various surfaces using both wind tunnel and field experiments. V_g values ranged from 8.6×10^{-5} to 2.4×10^{-3} cm/s.

Adams and Voillequé (Ada71) have shown that transfer of iodine to bromegrass is directly dependent upon the percentage of the leaf area composed by stomatal openings.

At the Jülich Nuclear Research Center (JNRC), Federal Republic of Germany, deposition of iodine has been investigated for several years. Vogt et al. (Vog71) report some results for preliminary experiments in a glove box. In three experiments they

found V_D equal to 13.6, 19.5 and 22.9 cm^3/g s for deposition of iodine on grass. In a second report (Voq73) average V_D values of four glove box experiments are 0.161 and 12.99 cm^3/gs for CH_3I and iodine respectively. Also results from 7 series of field experiments were reported. 6 of these dealt with grass, the last with clover. The results are discussed in connection with an empirical model

$$V_g = c \cdot B \cdot W \cdot u_*^2/u \ , \qquad (27)$$

where B is called a biological quality factor and c is a constant taking account of all other variables. In this work V_g is again found to be proportional to vegetation density, $V_g = 0.0045 \cdot W$, W is in g/m^2. When c and B is combined in one parameter A (cm^2/g), the 7 series of experiments are put into 3 groups with average values of A 66±4, 34±5 and 10±3 cm^2/g for normally growing clover, rapidly growing grass and slowly growing grass, respectively. The measured V_g values show no correlation with V_g values predicted from diffusion theory.

Further experiments from JNRC are reported by Vogt et al. (Vog74). V_D for CH_3I to grass has been found to be to orders of magnitude less than V_D for iodine. The experimental results are discussed on the basis of the following expressions

$$V_g = c \cdot B \cdot W \cdot u_* \cdot F \qquad (28)$$

or

$$V_D = c \cdot B \cdot u_* \cdot F = A \cdot M \ , \qquad (29)$$

where F is the relative humidity, $M = u F$ is called the meteorological parameter and the proportionality constant c contains as before all other parameters except B which influence the deposition but whose qualitative effects are unknown up to now. The values of W, u and F can be measured directly. $A = c \cdot B$ must be determined from the experimental results. A is found to vary from around 2 cm^2/g in spring and autumn to around 10 cm^2/g in summer. When comparing the highest values of A for grass with those for clover, it is found that the deposition velocity of iodine

onto clover, under equal meteorological conditions and with the same dry weight, is more than twice as high as that of iodine onto grass. The last report from JNRC (Hei76) contains additional experimental results, but no contributions to further understanding of the dry deposition of gases.

One of the many papers dealing with deposition of non-radioactive pollutants should be mentioned here. Fowler (Fow78) observed that when plants were beginning to age in late summer, deposition velocities for SO_2 were unexpectedly larger at night than during the day. This cannot be explained by stomatal opening because the stomata are permanently closed at this growing stage. The larger values occurring at night are explained by the presence of a layer of dew on the foliage. This might influence deposition of iodine, since iodine is also very soluble in water.

5.1.1.3. Conclusions. When estimating the dry deposition of radioactive gases the first thing to be aware of is that material cannot be deposited faster than it can be transported to the ground by the turbulent flow. This means that the minimum resistance or the maximum deposition velocity is given by equation (17) or (16) respectively. Laminar sub-layer resistance for grass can be evaluated in terms of B^{-1} found to be approximately 8 for ThB. Canopy or stomatal resistance is impossible to calculate directly. With the extensive experimental material presented here it is possible to give reasonably accurate deposition velocities for iodine to vegetation in many different situations. It is very unlikely that V_g for iodine vapour will exceed 1.0 cm/s. V_g for methyl iodide is two orders of magnitude less than V_g for iodine.

5.1.2. Dry deposition of particles

5.1.2.1. Introduction. Radioactive effluents may be discharged in particulate form or may also be absorbed more or less effectively on particles present in the atmosphere. The nuclei in the atmosphere upon which radioactive nuclides may be absorbed originate in many ways. They can be natural or man made. Natural aerosols can originate from e.g. sea spray, sand and dust blown from the grown or from volcanic eruptions, or they can be spores and pollen grains. Sources of man made aerosols are combustion processes, debris from nuclear weapon tests etc. Raindrops are not considered particles in this context.

Dry deposition of particles on vegetation surfaces takes place by mechanisms which are radically different from those by which vapours are transfered. Particles are deposited on plants by sedimentation and impact. The relative importance of these two processes depends primarily on particle diameter. Particles may be retained on impact, they may rebound or they may be retained temporarily and then subsequently removed. To distinguish the last two cases, the expressions bounce off and blow off will be used.

All particles are subject to the force of gravity and to a drag force dependent on the motion of the particle relative to that of the air. Electrostatic and thermal forces are very unlikely to be important for the deposition of aerosols in the open air (Chm60).

If a particle of diameter s moves with the velocity V_p relative to the air, the particle Reynolds number is

$$Re = s \cdot V_p \cdot \nu^{-1} \quad , \tag{30}$$

where ν is the kinematic viscosity of air (0.15 $cm^2 cm^{-1}$ at 20°C). For a particle with s = 10 µm and V_p = 1 cm/s, Re is approximately 0.01.

When Re is small (less than 0.1) Stokes' law applies giving the drag on spherical particle as

$$\text{drag} = 3 \cdot \pi \cdot \rho \cdot \nu \cdot s \; V_p \; , \tag{31}$$

where ρ is the density of air ($\rho = 0.0012$ gcm^{-3}). By setting the drag equal to the gravitational force minus the buoyancy force, the terminal velocity V_s of the spherical particle with density ρ_p is given by

$$\frac{1}{6} \pi \; s^3(\rho_p - \rho)g = 3\pi\rho\nu s V_s \; , \tag{32}$$

$$V_s = \frac{s^2 \cdot g}{18\nu} \; \frac{(\rho_p - \rho)}{\rho} \; . \tag{33}$$

Since ρ is usually much less than ρ_p, V_s may be expressed as

$$V_s = g \cdot t_r \; , \tag{34}$$

where

$$t_r = \frac{s^2}{18\nu} \frac{\rho_p}{\rho} \; . \tag{35}$$

t_r is called the relaxation time of the particle, and it is the time it takes for a falling particle, released from rest, to reach its terminal velocity. The drag on a non-spherical particle is also proportional to the velocity, still provided Re is small. The drag may then be expressed as

$$\text{drag} = 3\pi\rho\nu s V_{ps} \alpha \; , \tag{36}$$

where s now is the diameter of the sphere of equal volume, V_{ps} is the particle terminal velocity, and α is a constant called the dynamic shape factor, which depends on the shape but not the dimensions of the particle. Experimental values of α are known for different shapes.

Gregory (Gre61) has defined the trapping or impact efficiency of an obstacle, e.g. a leaf, as the amount of trapped matter per unit area per time unit divided by the flux. This can be shown to be equivalent to

$$V_g = u \cdot C_p \ , \tag{37}$$

where C_p is the impact efficiency. Deposition due to impact can only be treated theoretically for sticky obstacles of regular shape, e.g. sticky cylinders. The practical usefulness of impact theory for estimation of deposition velocities of particles to vegetation is very limited because of the irregular shapes of natural surfaces.

From theory the minimum deposition velocity of particles to the ground will be the terminal or sedimentation velocity, V_s. In still air it will be equal to the total deposition velocity, V_g. At wind speeds greater than zero the total deposition velocity, V_g, will have a contribution from impact, V_i,

$$V_g = V_i + V_s \ . \tag{38}$$

V_i will still be limited by the fact that material cannot be deposited faster than it can be transported to the ground by the turbulent flow,

$$\max V_g < \frac{s^2 g \rho_p}{18 \nu \rho} + u_* \ \frac{u_*}{u} \ . \tag{39}$$

5.1.2.2. Review of experimental data. Deposition of particles on vegetation has been studied in laboratory and field experiments. Fallout from nuclear ewapon tests and of other substances found in the atmosphere provide further information. Most of the experimental values of deposition velocities available are for grass surfaces. Some typical results of measurements of deposition velocities for particles are shown in table 3.

Table 1. Some typical results of measurements of V_g for particles.

Reference	V_g (cm s^{-1})	Surface	u_* (cm s^{-1})	Particle diameter (um)	Type of experiment	Comments
Chamberlain (1967)	0.7 - 3.5	grass	12 - 94	32	field	
	1.4 - 4.5	grass and soil	17 - 94			
	0.4 - 5.6	filter paper	12 - 94			
	2.9 - 9.2	grass	18 - 183		wind tunnel	
	4.9 - 35.7	sticky artificial grass	28 - 146			
	0.033 - 4.9	grass	70	0.08 - 2		
	0.012 - 13	sticky artificial grass				
Wilson (1967)	0.8	alfalfa			field	mean value for a 25 days period.
Peirson (1973)	0.1 - 0.7	filter paper		0.1 - 10	field	study of trace elements
Belot (1974)	0.007 - 40	pine	1 - 10	2 - 10	wind tunnel	
	0.001 - 35	oak				
Clough (1975)	3.4	moss	37	30	wind tunnel	
	7.3		87			
	11.1	wet moss				
	0.35 - 1.45	moss	37	3.5 - 6.5		
	0.6 - 0.66	wet moss		4.5 - 6.5		
	0.75	moss		3.0		
	0.08	grass	70	0.8		
Horbert (1976)	0.1	grass	27	4	field	
	0.24	clover				
	0.035	bare soil				
Wesely (1977)	0.12 - 1.18	grass	5.5- 19.5	0.05 - 0.1	field	
Jones (1979)	0.012 - 0.39	different surfaces	14 - 38	2 - 6	field	

In the very extensive study by Chamberlain (Chm67) lycopodium spores tagged with radioactive markers, mean diameter = 32 µm, as well as aerosols of various smaller particle sizes have been used in field and wind tunnel experiments to determine deposition velocities to grass and other surfaces. Particle size has an important influence on aerosol behaviour. In figure 5.2 the variation of deposition velocity with particle size is shown.

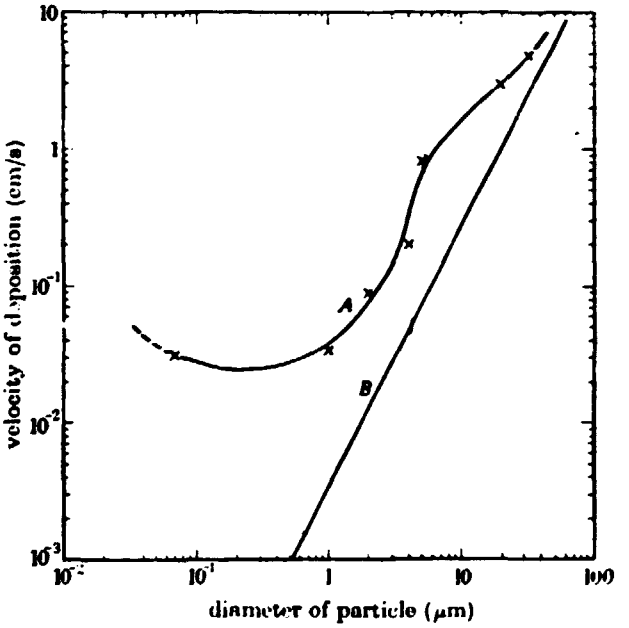

Fig. 5.2. Velocity of deposition of particles to grass in wind tunnel experiment. u_* = 70 cm/s. A, experimental results; curve B represents the terminal or sedimentation velocity. (Chm67).

Sedimentation dominates for large particles. For diameters between 0.1 and 1 µm, the deposition velocity has a minimum. For sub-micron particles V_g increases again due to the increasing contribution of Brownian motion. The dependence of V_g upon particle size is also shown in figure 5.3, but for three different values of the friction velocity. The same variation is

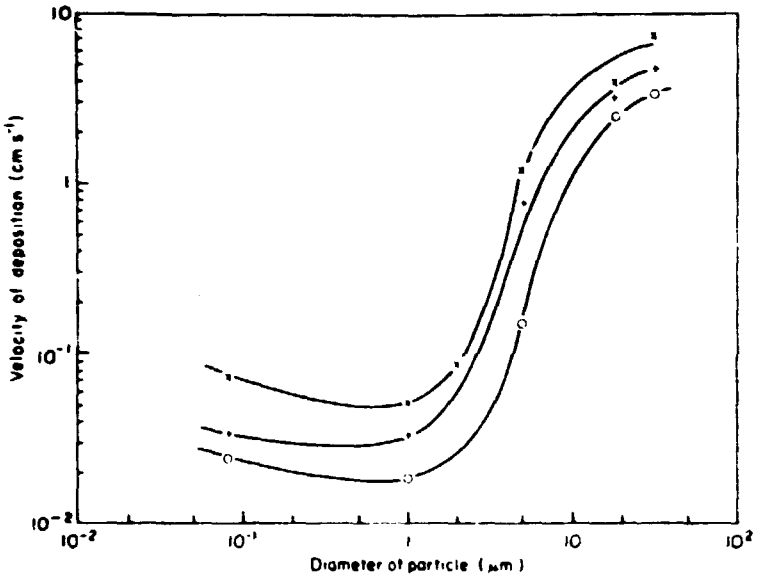

Fig. 5.3. Deposition of particles to grass in wind tunnel experiment for three different u_* values. (x) u_* = 140 cm/s, (+) u_* = 70 cm/s, (0) u_* = 36 cm/s. (Chm75b).

observed and V_g varies by 2 orders of magnitude with particle sizes from 1 to 30 μm. V_g was found to be almost equal to the terminal velocity, V_s, for very low windspeeds, but at higher speeds, deposition by impact becomes more important. The effect of surface structure and stickiness was investigated. More spores were deposited on wet than on dry surfaces at high friction velocities, but there was little difference at low friction velocities, where sedimentation is more important than impaction. The role of stickiness or wetness is straightforward: once in contact particles are less likely to leave. Chamberlain's results also show, that stickiness of the surface is an important factor in catching larger particles, but for particles less than 5 μm, bounce off is much less likely to occur irrespective of the degree of stickyness and the presence of hairs and other small irregularities on the leaves, appears to be more important.

Peirson et al. (Pei73) determined some 30 elements quantitatively in aerosol samples. The dry deposition velocities ranged from 0.1 to 0.7 cm/s. For Cs and Sr the data were incomplete, and there were no measurements of the particle size distribution. The aerosol was characterized by a mean diameter d ~ 0.5 µm derived from the measured V_g values. It is, however, unlikely that a natural aerosol can be represented by a single value of d.

Belot and Gauthier (Bel74) measured transfer rates for 2, 5 and 10 µm particles to small branches of pine and oak trees in the laboratory. The measured deposition velocities varied over several orders of magnitude for a variation of the wind speed from 1 to 10 m/s for all three particle sizes. Again it was found that deposition velocities for the smaller particles were highest for fibrous leaves.

Clough (Clo75) investigated the transport of particles of various sizes to moss and rye grass in a large wind tunnel. The experiments were set up to see if moss bags could be used as monitors of dry particulate input to extended crop and soil surfaces. The usual effects of particle size and wind speed on deposition velocities of particles were also confirmed in this study.

JNRC has carried out a number of field experiments concerning the deposition of radioactive-labelled copper sulfate aerosols of various size onto vegetation and other boundary surfaces (Hor76). Particle size distribution was measured with an aerosolspectrometer. Representative values of V_g for 4 µm particles and $u_* = 27$ cm/s are shown in table 3.

Wesely (Wes77a) made eddy-correlation measurements determining the deposition velocity as

$$V_g = \overline{U'X'}/\overline{X} \quad , \tag{40}$$

where U'X' is the covariance of particulate concentration, X, with vertical wind speed U. One of the main reasons for a great

uncertainty of 40% is probably the measuring of wind speed in the height of 5 m with an anemometer with a spatial response of 2 m. The effect of atmospheric stability on deposition velocities could not be determined with great accuracy, but the results imply that the diurnal variation of atmospheric stability should be taken into account in order to parameterize deposition processes realistically.

Jonas (Jon79) has tried to correlate measured V_g values with vegetation density, friction velocity u_*, and particle diameter, s, in the equation

$$V_g = c\ B\ W\ u_*\ s\ , \qquad (41)$$

where c is a constant and B is a biological parameter. Correlations with u_* and W were established, but not with s.

Wilson et al. (Wil67) measured the deposition velocity for Cs-137 on sheltered and unsheltered alfalfa. He draw the same conclusion as May (May58): dry deposition is not negligible during rainfall.

5.1.2.3. Conclusions. The variation of particle dry deposition velocity with particle size and friction velocity is very well determined. Minimum and maximum V_g values can be given for different conditions. The major uncertainty in estimating deposition of an aerosol to natural surfaces is probably the particle size distribution within the aerosol. Aerosol size spectra have been published, see e.g. (Chm75b, Sli75). Figure 5.3 probably gives very realistic particle deposition velocities.

5.2. Precipitation scavenging

5.2.1. General description

Wet deposition is more correctly denoted *precipitation scavenging*. The literature has all the aspects of the very non-uniform terminology that exists in this field. The text below will

contain definitions as suggestion for a more stringent and consistent terminology. Figure 5.4 illustrates the main concepts of precipitation scavenging. Precipitation scavenging is defined

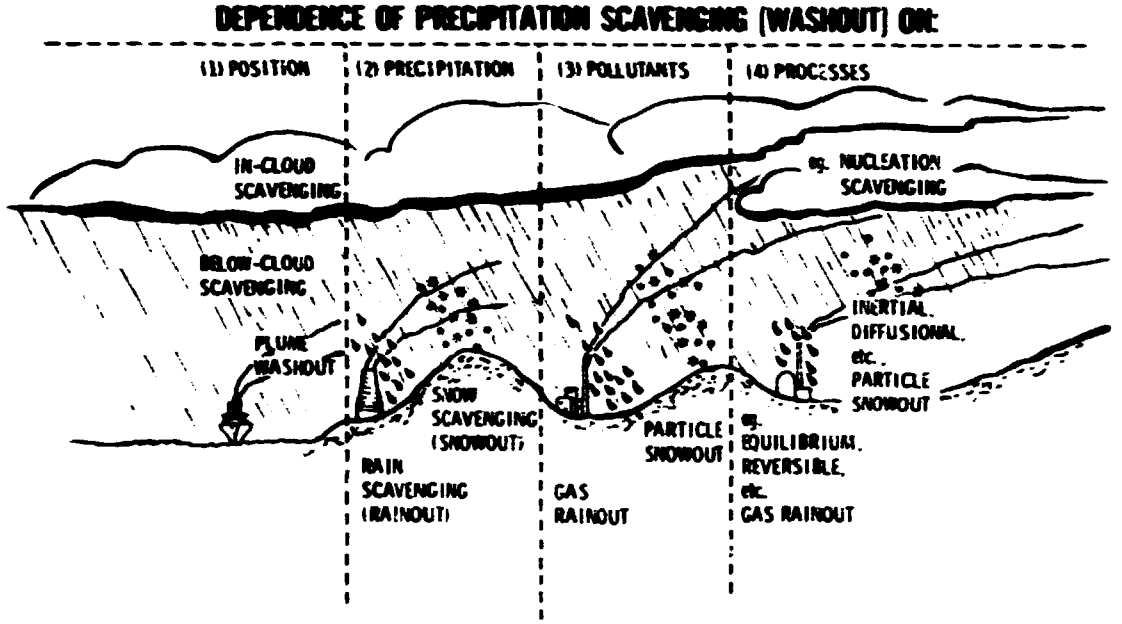

Fig. 5.4. A schematic illustration of some basic concepts in precipitation scavenging. After (Sli79).

as removal of any material from the atmosphere to the earth's surface by various types of precipitation. That is: liquid or frozen atmospheric water called hydrometeorites, with a gravitational terminal velocity about 10 cm/s. The words *material* and *matter* denote both particulates and gases. "Washout" is often used as an abbreviation for precipitation scavenging.

The three first sections of figure 5.4 illustrate the possible situations which determine relevance of the various processes, as shown in the fourth section.

Fig. 5.4 (1): Precipitation scavenging can be considered as a function of where the processes take place. Below-cloud scavenging denotes washout beneath the visible cloud, which is the source of the precipitation. In-cloud cavenging means scavenging of material within the visible cloud, which results in deposition on the earth's surface. Unfortunately *rainout* is often used to mean in-cloud scavenging. *Rainout* is more logically used to describe rain scavenging. In-cloud scavenging is important in deposition of airborne debris from atmospheric nuclear weapon testing. In a convective weather situation material released at ground level may be swept up into the clouds. The distinction between in- and below-cloud scavenging may not be easy with respect to experimental research, since some of the processes are the same with only the size distribution of the hydrometeorites being different. At ground level it is difficult to distinguish between in- and below-cloud scavenging. Only one experimental study of in-cloud scavenging (Sli79) has been found in the literature reviewed here. In-cloud scavenging will be discussed in the final part of this chapter (5.2.1).

Fig. 5.4 (2): The type of precipitation, rain or snow, is of cource important. One would expect that snow and ice crystals should be more efficient scavengers than water drops, because of their larger surface-to-volume ratio. Snow scavenging is also called "snowout" and rain scavenging is corespondingly denoted "rainout". Few studies have been devoted to snow scavenging. One reason may be the large variety of snow types; another the lack of even approximate descriptions of flow fields about snow crystals.

Fig. 5.4 (3): The nature of the material to be scavenged has to be taken into account. If it is particulate the size distribution should be specified, and if it is gaseous the diffusivity and solubility should be specified. With increasing time and distance from a source these characteristics are modified by agglomeration, fragmentation and by attachment and adsorption onto "natural" aerosols. The "natural aerosol" does not have well defined parameters, the size distribution varies with elevation, source and meteorology. Junge (Jun63) presents model size distributions, that can be used in the absence of measurements.

Fig. 5.4 (4): The scavenging is a sum of a bewildering number of processes: e.g., Brownian motion, thermophoresis, diffusiophoresis, turbulent inertial interception, and gravitational capture; see e.g. (Pru78). The scavenging processes may be considered as material being exposed to moving precipitation elements with some chance of collection. Consequently, scavenging can be described by an exponential function:

$$X(t) = X(0) \cdot \exp(-\Lambda t) \qquad (40)$$

where X(0) the concentration at time zero, X(t) is the atmospheric concentration of material at time t, Λ in units of time^{-1} is called the washout or scavenging coefficient. Λ denotes the fraction of material removed per unit time. The basic assumption is implicitly that the fraction of material removed per unit time is independent of the amount of material present. Usually the exponential description will be true but if there are too many particles which act as cloud-droplet or ice-crystal nuclei then their presence could influence the precipitation rate, and some gases can saturate the cloud droplets and then only a certain amount, not a specific fraction, of the gas will be removed.

In some references (McM79, Gyl80, HÜ80) a "Λ" is expressed as $\Lambda = c\ p^a$, where c is a constant in s^{-1}, p the precipitation rate in mm h^{-1} and a a number between 0 and 1. This type of relationship is a consequence of the observation that drop size distribution is dependent on the rainfall intensity; a higher intensity gives larger drops.

Numerous measurements of raindrop size distributions have been made, and several empirical equations have been fitted to them. Rain spectra have peak frequencies betwen drop diameters of 0.5 and 1.0 mm. The distributions given by Marshall and Palmer (Mar48) and Best (Bes50) have been most widely accepted and used. Rain spectra sampled by Kelkar (Kel59) are shown in figure 5.5. In this figure it is seen how raindrop size distribution varies with rainfall rate. For a rainfall rate of 0.2 mm/h 30% of the drops have diameters greater than 0.4 mm, but for a rainfall rate of 8.6 mm/h 30% of the drops have diameters

greater than 2 mm. It is also seen that when the rainfall rate is high, 8 mm/h or greater, the raindrop size distributions do not vary very much. Users of rain spectra should consider the effects of evaporation as the rain falls to the ground.

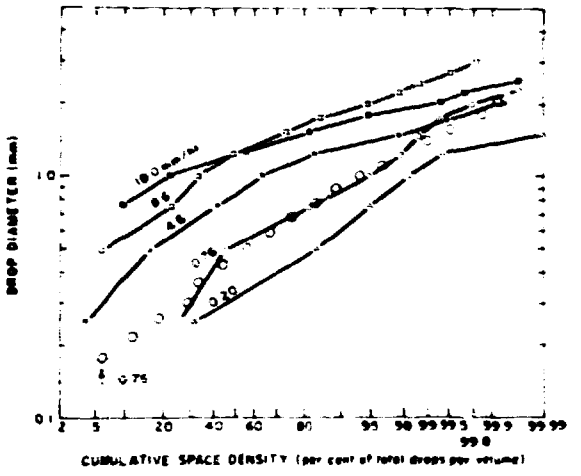

Fig. 5.5. Selected rain spectra measured by Kelkar (Sla68).

In contrast to what is a popular belief among people, it is raining or snowing less than 10% of the time in most European countries. From precipitation statistics (Gyl80) covering a 5 and 7 years period in Sweden and Denmark respectively, it is seen that there is precipitation (at a rate greater than 0.1 mm h^{-1}) 7.9% and 7.2% of the total time. The same satistics show that in 92.5% and 93.5% of the total precipitation time, the precipitation rate is less than 2 mm h^{-1}. 85.7% of the total time precipitation is during weather situations characterized by Pasquill categories D and E. It should also be pointed out that precipitation at a rate greater than 1 mm h^{-1} during a very stable weather situation, Pasquill category F, has only occurred 0.0076% of the total time.

It is generally believed (Jun63, Mah67, Bur68) that in-cloud scavenging is relatively more efficient than below-cloud scavenging. The conclusion was based on analyses of atmospheric bomb debris in precipitation. The only direct experimental test of the relative importance of below versus in-cloud scavenging has recently been performed by Slinn et al. (Sli79). This study gave evidence of in-cloud scavenging being as efficient as below-cloud scavenging. For a consequence-assessment model this means that in situations with rain it probably does not make much sense to distinguish between in-cloud and below-cloud scavenging. However, in some situations initially with no rain in-cloud scavenging can affect the local deposition. Firstly, when a plume diffuses beyond the condensation level it may cause condensation and cloud formation. Hence it can cause precipitation such that both in-cloud and below-cloud scavenging will occur. Second, when a plume reaches into the condensation level the additional condensation nuclei which it contains may compete with nuclei present, such that no precipitation elements will be formed. Hence it can prevent precipitation to the extent that no scavenging will take place. In this situation the dry deposition will be lower than in situations without in-cloud scavenging.

More details can be found in Engelmann and Slinn (Eng70), Semonin and Beadle (Sem77), Pruppacher and Kleft (Pru78), and Slinn (Sli80).

5.2.2. Precipitation scavenging of gases

5.2.2.1. Introduction. When dealing with precipitation scavenging of gases, the gases should be divided into two categories: those that are highly reactive towards water, e.g. bromine and iodine, and those less reactive that form simple solutions in water, e.g. CH_3I and CO_2.

For highly reactive gases rain scavenging coefficients may be calculated from the equation (Chm53b):

$$\Lambda_g = \int_0^\infty Sh\ D\ \Pi\ s_h\ N(s_h)\ ds_h \qquad (42)$$

where D is the molecular diffusion coefficient and $N(s_h)$ the number of drops of diameter s_h in a unit volume of air. Sh is the Sherwood number, which for spherical drops is approximately (Ran52):

$$Sh = 2.0 + 0.6\ Re^{1/2}\ Sc^{1/3}\ . \qquad (43)$$

Here, Re is $s_h v_t/\nu$ and Sc, the Schmidt number, is ν/D, where v_t is the drop terminal velocity and ν is the kinematic viscosity of the air. Sh D in (42) can be regarded as a kind of collection effeciency for highly reactive gases.

5.2.2.2. Review of experimental data. Very few studies on gas scavenging were found in the literature reviewed. This is probably because particle scavenging is considered more important.

Engelmann et al. (Eng66a) have calculated scavenging coefficients for highly reactive gases for five different rain spectra. In figure 5.6 instead of the calculate Λ_g's, Λ_g/D is shown, because Λ_g (42) is almost linear in D. Engelmann et al. (Eng66a) recomend use of the Kelkar rain spectra (Kel59). In figure 5.7 predicted Λ_g values for bromine and iodine using Kelkar rain spectra, together with Λ_g values measured by Engelmann et al. (Eng66a) and Engelmann and Perkins (Eng66b). The measured values spread over three orders of magnitude. However, measured Λ_g's for bromine fit the theoretical predictions. The tremendous difference between scavenging iodine and bromine can be attributed to the different chemical and physical properties of these elements towards water, e.g. bromine is a hundred times more soluble in water than iodine.

Engelmann and Perkins (Eng66b) used iodine released from a process plant in their study. The washout coefficients from this study (Eng66b) are high compared to those of other investigations. This can be explained by presence of an amount of water vapour in

the plant exhausts, sufficient to produce a cloud of water droplets. If the released iodine is inside water drops before scavenging, the washout rate would be equal to that of the droplets. This can explain that the values of measured washout coefficients are high. Hence, release conditions can play a very important role when evaluating washout.

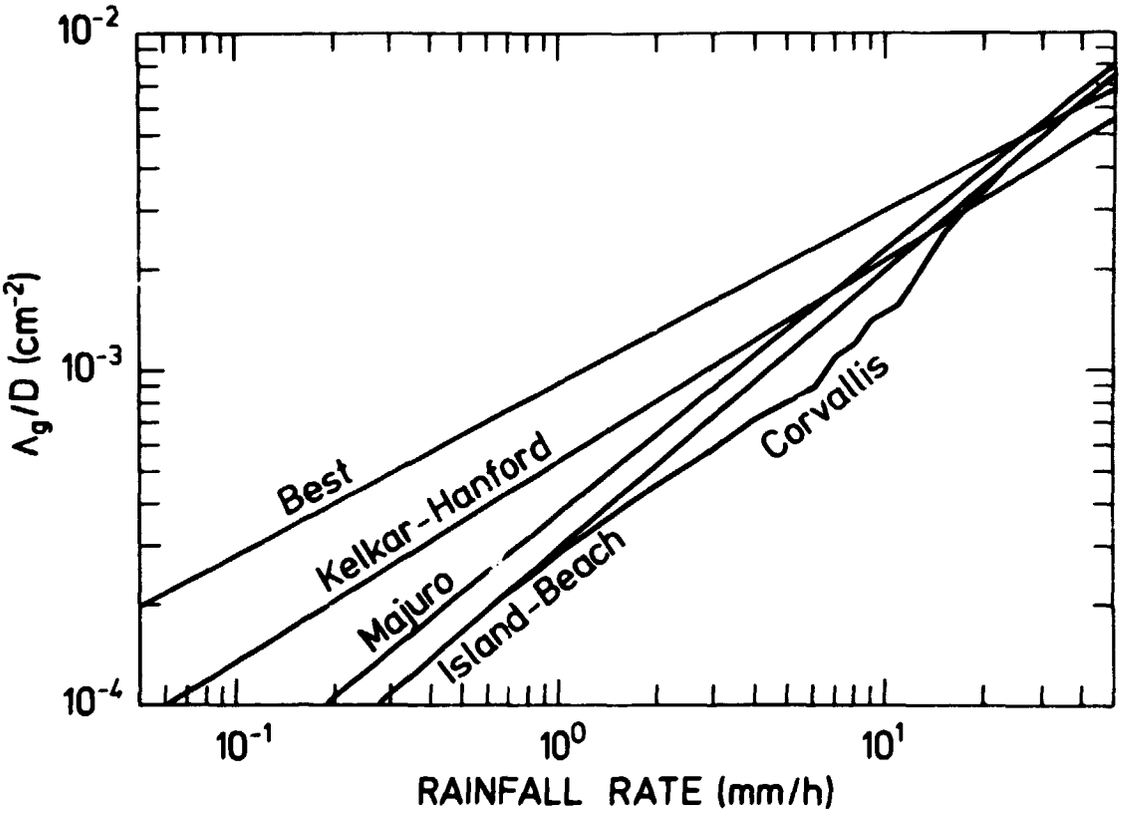

Fig. 5.6. Scavenging coefficients for soluble gases divided by diffusivity, predicted from different rain spectra (Eng66b).

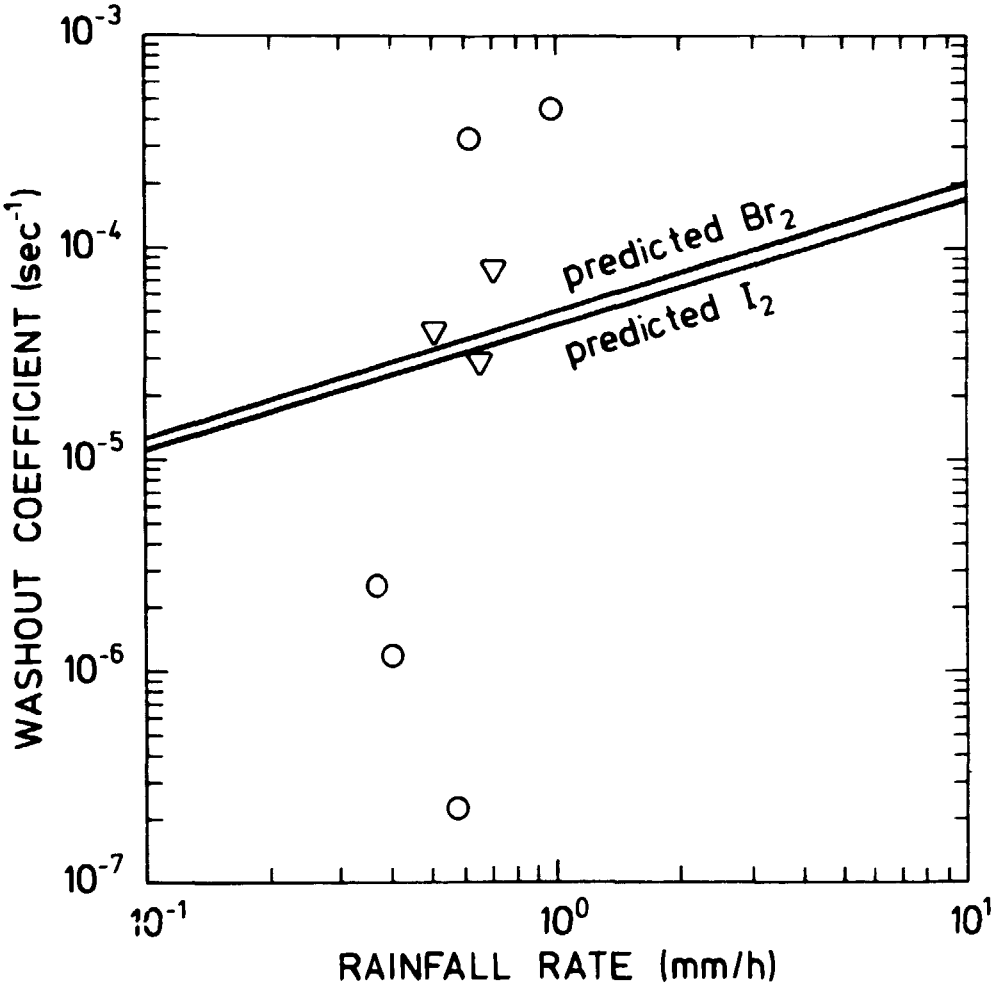

Fig. 5.7. Measured scavenging coefficients for bromine (∇) and iodine (O) compared with predictions (Eng66b).

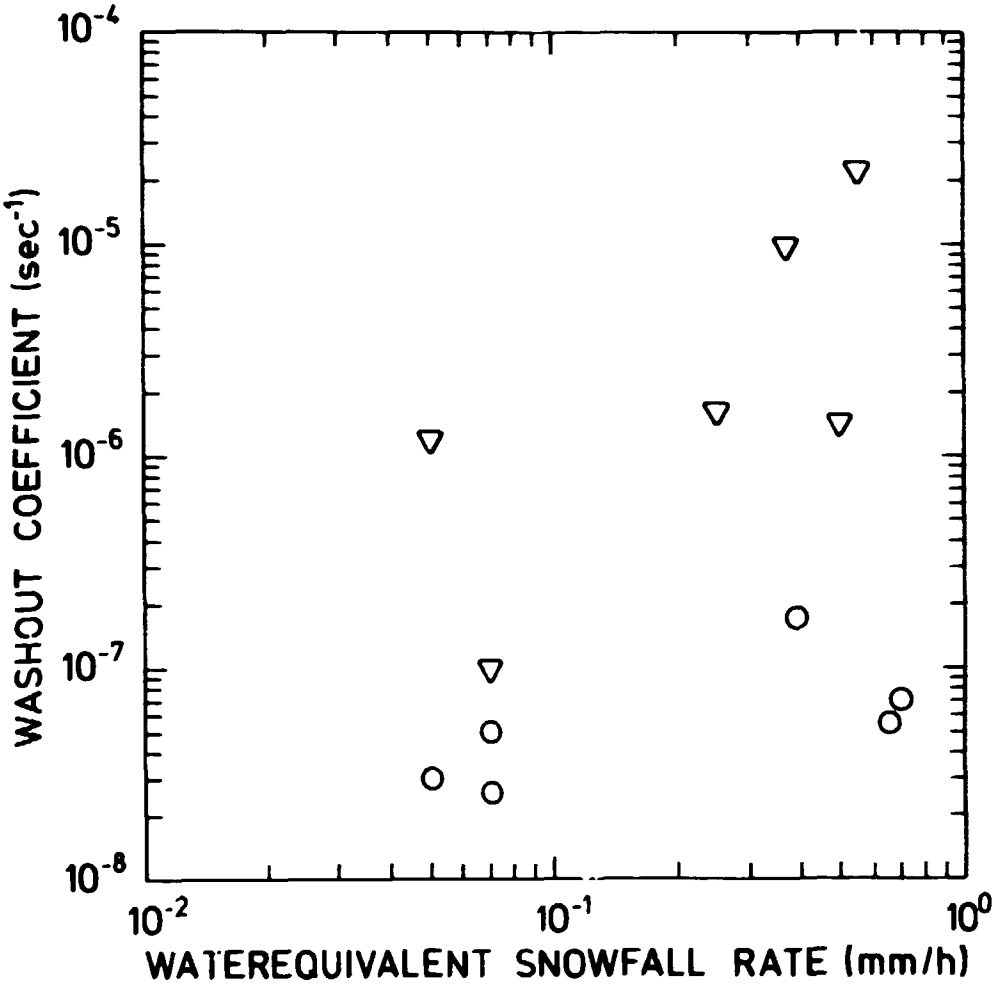

Fig. 5.8. Measured scavenging coefficients for bromine (Δ) and iodine (O) versus snowfall rate. (Eng66b).

Data given by Engelmann and Perkins (Eng66b) show that snowout can be calculated as a linear function of the waterrequivalent precipitation rate. Snowout of bromine and iodine have also been measured by Engelmann et al. (Eng66a), see figure 5.8.

Bromine is scavenged approximately two orders of magnitude more effectively than iodine. Measured Λ_g's for snowout are lower than for rainout, but there are large variations in the results. Since the number of snow crystals and the crystal size distributions change with the precipitation rate this spread in data is not surprising. The coefficients being lower for snow than for rain, support the importance of solubility and reactivity. Although large surface area and small terminal velocities of snow crystals may increase the scavenging of gases by snow, this is not able to compensate for the lower absorption rates at ice surfaces.

Experiments performed in connection with spray systems installed inside reactor containments for safety reasons have given a lot of data on iodine scavenging (Nucl. Technol. 10, 405-553 (1971)). However, these experiments were carried out using much higher iodine concentrations than would be relevant to atmospheric situations. Moreover, the sprays often contained chemical additives.

5.2.2.3. Conclusions. Data from experiments performed with reactor containment spray systems cannot be used for evaluation of wet atmospheric deposition.

Snowout coefficients for bromine and iodine show large variations and are approximately one order of magnitude lower than the corresponding rain scavenging coefficients.

The rainout coefficients for organic forms of iodine were found to be 1% of those of elemental iodine (Eng66a), this is consistent with the difference in reaction rate towards water.

The only measured Λ_g values relevant for iodine are those of Engelmann et al. (Eng66a, Eng66b), and obviously more work needs to be done is this field. Λ_g for iodine vapor can be set to $10^{-6}s^{-1}$ for precipitation intensities of 5 mm/h and less. This value is an "order-of-magnitude" recommendation.

5.2.3. Precipitation scavenging of particles

5.2.3.1. Introduction. When a raindrop or snowflake moves it sweeps out a volume of air. Particles in this volume of air will tend to follow the air. Inertia will cause some fraction of the particles to intersect with the raindrop or snowflake. This fraction is called the target or collision efficiency.

Some particles may bounce off the raindrops or snowflakes and they are not collected even though they have had contact. The fraction of those making contact and remaining with the raindrop or snowflake is called the retention effeciency.

The product of target efficiency and retention efficiency is the collection efficiency, E. The collection efficiency can as a first approximation be regarded as a function of particle diameter, s_p, and hydrometeor- (rain-drop or snow-flake) diameter s_h

$$E = f(s_p, s_h) \quad . \tag{44}$$

Some information regarding target efficiencies is available in the literature (Lan48). Very little is known about retention efficiencies. A likely assumption is that of perfect retention.

The interesting result of adding the effects of various scavenging mechanisms was first obtained by Greenfield (Gre57). His model gave scavenging coefficients showing a strong broad minimum for aerosol particles between about 0.1 and 1.0 μm radius. This minimum is often referred to as the "Greenfield gap" or the "scavenging gap".

Slinn (Sli77a) has suggested a semi-empirical expression for the collection efficiency. This expression accounts for particle Brownian motion, interception and inertial capture and the collision efficiency for two different drop sizes is shown in figure 5.9 as a function particle radius.

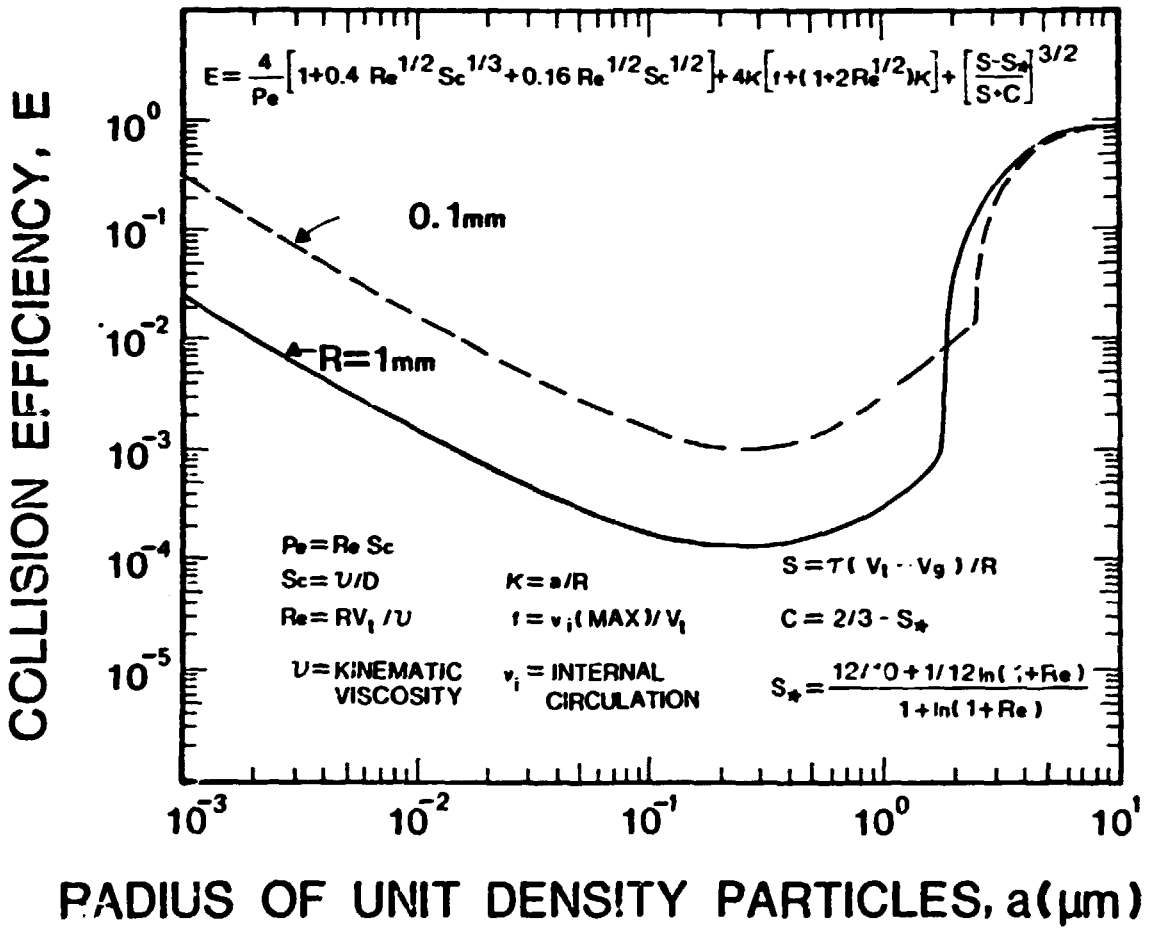

Fig. 5.9. The particle/drop collision efficiency is shown evaluated for two drop radii. (Sli80).

When E is a known function of s_p and s_h, then the washout coefficient for a specific aerosol particle diameter, $\Lambda(s_p)$, can be calculated by integrating over all drop sizes:

$$\Lambda_p(s_p) = \int_0^\infty N(s_h) v_t(s_h) \frac{\pi}{4} s_h^2 E(s_p, s_h) ds_h \ , \tag{45}$$

where $N/s_h)$ is the size distribution function for hydrometeors and $v_t(s_h)$ is their terminal velocity. The rainfall rate can be expressed as:

$$p = \int_0^\infty N(s_h) v_t(s_h) \frac{4}{3} \pi \frac{1}{8} s_h^3 \, ds_h \tag{46}$$

When comparing (45) and (46) one can suggest the following approximation to (45) for the rainout coefficient, Λ_p^r:

$$\Lambda_p^r (s_p) \approx \frac{p(x,y,z,t)}{s_h^m} E(s_p, s_h^m) \ , \tag{47}$$

where s_h^m is the volume-mean drop diameter, which is usually (Man71) related to rainfall rate p by:

$$s_h^m = 0.7 \text{ mm } p^{1/4} \tag{48}$$

The approximation for the snowout coefficient, Λ_p^s, is not quite as good as the one introduced for the rainout coefficient, Λ_p^r. Slinn (Sli77b) has also suggested an approximation for Λ_p^s:

$$\Lambda_p^s (s_p) \approx g \frac{\rho_w}{\rho_a} \frac{p(x,y,z,t)}{\langle v_t^2 \rangle} E(s_p, s_h^m) \tag{49}$$

where s_h denotes the diameter of the snowflake or the ice crystal, p is the rainwater equivalent precipitation rate, and $\langle v_t \rangle$ is the average settling speed of the snowflake. Theoretically scavenging coefficients for an aerosol can be calculated by integrating (45) over the aerosol particle size distribution.

5.2.3.2. Review of experimental data. Chamberlain (Chm53a) has used Langmuir's target efficiencies (Lan48) and Best's rain spectra (Bes50) to calculate washout scavenging coefficients for various rainfall rates and particle sizes. These calculated washout coefficients are shown in figure 5.10 for different values of $s_p^2\rho_p/4 = a^2\rho_p$ where a is the radius and where ρ_p is the density of the particle. It is seen A_p is almost proportional to both rainfall rate and particle diameter.

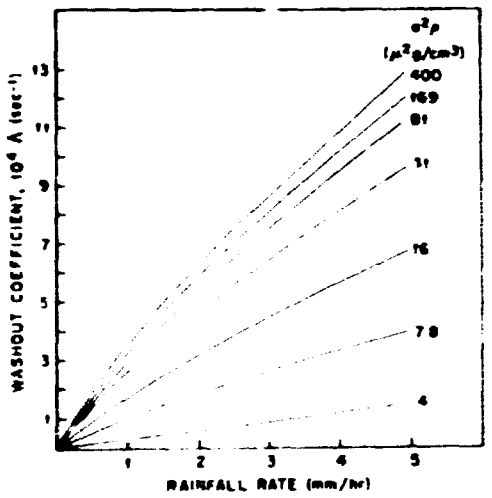

Fig. 5.10. Washout coefficients for unit density particles vs. rainfall rate and $a^2\rho$, where a and ρ are the radius and density of the particles. (Chm53a).

Experimental work can be done as follows. Firstly the collection efficiency for each dropsize on each particle size is measured; secondly the size of falling drops is measured; thirdly the collection efficiencies are integrated over all raindrop and particle sizes. However, the procedure is very tedious. In figure 5.11 some results from such work on zinc sulfide particles are shown (Eng65).

The measured collection efficiencies become quite small as the particle size decreases. Theoretically E goes to zero with

decreasing particle diameter. Unfortunately Engelmann (Eng65) made no measurements on natural washout of submicron particles.

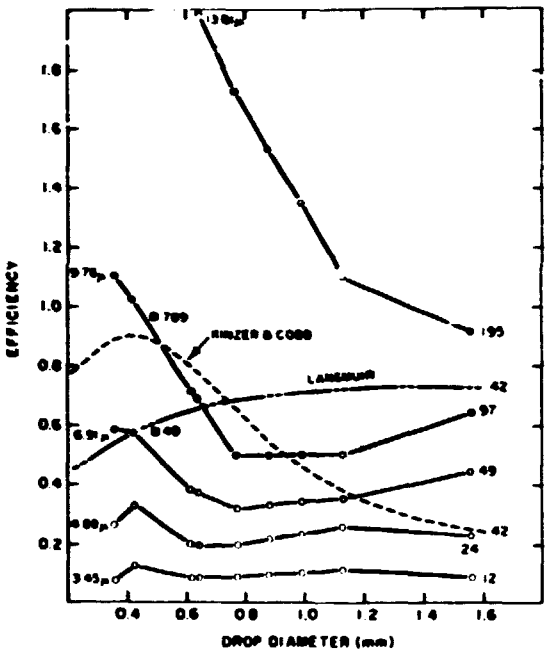

Fig. 5.11. Experimental curves for zinc sulfide particle collection efficiencies vs. drop diameter for various $s_p^2\rho_p$ (μm^2 g/cm^3), with Langmuir (Lan48) and Kinzer and Cobb (Kin58) data entered as dashed lines (Eng65).

Washout by snow could be expected to be more effective than for the water equivalent of rain, because slow "feathery" snowflakes have larger surface areas than the equivalent waterdrops.

Very few field measurements of washout of aerosols by snow have been done. One reason might be the large variety of crystal types that exist. Results from the study be Graedel and Franey (Gra75) indicate snow scavenging to be 28-50 times more effective than water equivalent rain scavenging.

McMahon and Denison (McM79) have compiled comprehensive lists of field measurements of washout coefficients and washout ratios. The scavenging coefficients obtained vary over three orders of magnitude. In fig. 5.12 results from Radke et al. (Rad74) have been plotted together with values for which the particle sizes are known. As mentioned earlier the Λ's depend in part on precipitation intensity. Figure 5.12 is for a mean rainfall rate of 5 mm/h. In figure 5.12 the scavenging coefficients vary over two orders of magnitude depending on particle size, and have two distinct minima around particle diameters of 0.1 and 1.0 μm. However, it is not clear from the reference (McM79) how the solid curve has been drawn. Recently Radke et al. (Rad80) report measurements of scavenging efficiencies as a function of particle size. They did not find the hump in the "scavenging gap" in Figure 5.12, and they found the "scavenging gap" to be much narrower.

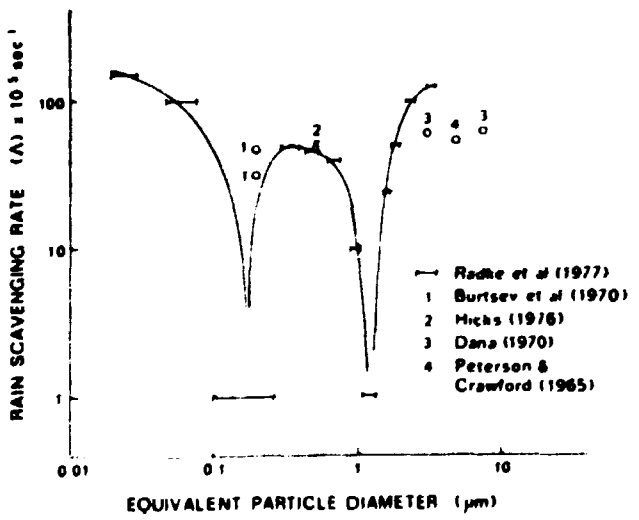

Fig. 5.12. Relationship between rain washout coefficients and particle size. (Rad74).

5.2.3.3. Conclusions. The washout coefficient, A_p, depends mainly on particle and raindrop size distributions. In figure 5.10 is calculated values are shown for different rainfall rates and different values of $s_p^2\rho_p/4$, where s_p is the particle diameter and ρ_p is the particle density.

In figure 5.12 is given the experimentally determined relationship between A_p and particle diameter for an approximate mean rainfall rate of 5 mm h^{-1}. From figure 5.12 an overall maximum A_p could be set to 0.001 s^{-1}. Pruppacher an Klett (Pru78) gives A_{max} in s^{-1} as:

$$A_{max} = 4.2 \times 10^{-4} E p^{0.79} , \qquad (50)$$

where E is a mean collision efficiency and p is the precipitation rate in mm h^{-1}. For E = 0.1 and p = 5 mm h^{-1} (50) gives $A_{max} = 1.5 \times 10^{-4}$. Pruppacher and Klett (Pru78) also point out that significantly higher scavenging coefficients are not expected in case of strong turbulence and higher rainfall rates. More than one order of magnitude accuracy should never be expected.

5.2.4. Washout ratios

A simplification and an alternative formulation for precipitation scavenging for both gases and particles uses the scavenging ratio, W, commonly called the washout ratio. The washout ratio is usually defined as the ratio of the material concentration in the precipitation at surface level, k_o (curies per volume of precipitation), to its average concentration in the air at surface level, X_o (curies per volume of air). Washout ratios are normally reported for a particular element or compound. It should be noted that some authors report washout ratios with dimensions such as m^3 cm^{-3}, kg air per litre water or m^3 l^{-1} and therefore readers may find washout ratios of different orders of magnitude.

The wet flux to the ground, W, can be written:

$$W = w\ p\ X_o\ , \tag{51}$$

and analogous to dry deposition a "wet deposition velocity" can be defined:

$$V_w = W/X_o = w\ p\ . \tag{52}$$

McMahon and Densison (McM79) have given a comprehensive list of washout ratios. Using typical values of $w = 1 \times 10^5$ and $p = 100$ cm y^{-1}, (52) gives a wet deposition velocity of approximately 0.3 cm s^{-3}.

Washout ratios do not by vary orders of magnitude like the scavenging coefficients. This apparent and deceptive difference in level of accuracy is easily understood. Washout ratios are just integrals of the scavenging coefficients. It is because the ratios are averages over a lot of parameters, over the precipitation element sizes, over the material's vertical distribution, over the particle sizes, over different chemical forms of the element, over different rainfall amounts, over different wind directions, over different types of storm over different sources etc., that they show very little variation. Using washout ratios is to use Natures own integrations.

When it comes to dose calculations after a hypothetical reactor accident, there is some difference of opinion regarding the usefulness of washout ratios for predictions of precipitation scavenging. The conclusion is that washout ratios can only be used in two situations. Firstly, when one is sure that Nature integrates in the same way. This means that washout ratios can be used at the same location, with the same weather situation etc. as when they were obtained. It should be remarked that washout ratios can vary by three orders of magnitude during a single rainstorm (Ros67). Secondly, washout ratios can be used for prediction of precipitation scavenging of routine releases, where long term averages are of interest.

6. RETENTION ON THE SURFACE OF VEGETATION

6.1. Introduction

The loss of material deposited on vegetation surfaces may be divided into two time intervals. Firstly, some part of the deposited material is immediately lost. Secondly, some part of the initially retained material is removed at a lower rate by different weathering processes. The loss rate in both cases depends on the deposit, the nature of the vegetation and the meteorological conditions before, during and after the deposition. The terminology in this area is quite ambiguous. The interception factor should be defined as the fraction of the total deposit to a surface initially remaining on the plants, where the rest of the deposit is transferred directly to the soil surface. Another very common name for the interception factor is initial retention factor.

Interception and retention factors should be used with great caution. It must be pointed out that it is not always necessary to use retention factors in risk assesment calculations. If the deposition velocities used have been derived from direct experiments on vegetation, the details of the experiments will determine whether or not interception or retention factors are required. If retention factors are used, the definition of these factors should be checked.

6.2. Retention during initial phase

Middleton (Mid60) has reported initial retention factors for strontium and caesium on wheat and cabbage of 8 to 60% during precipitation. It has been shown that 40 to 60% of iodine was retained on vegetation during wet deposition (Chm65). In field experiments Milbourn and Taylor (Mib65) obtained retentions ranging from 16 to 30% on the edible parts of different types

of herbage. Kirchmann et al. (Kir66) report less than 10% retention of strontium and caesium after spraying rye-grass with a density of 130 g dry weight per m^2.

Eriksson (Eri77) expresses the fraction retained, F, as

$$F = \mu W \ , \tag{53}$$

where μ is a retention coefficient in m^2kg^{-1} and W is the amount of grass dry matter per unit area in kg/m^2. He reports a retention of approximately 25% for caesium during wet deposition on pasture grass.

The retention values should not be considered as constants, as the values can be expected to vary with precipitation rate and with the properties of the material being deposited and with the morphology of the plants.

For dry deposition a 30% retention for 30 µm fallout particles on rye is reported in the review by Scott Russell (Rus65). Chamberlain (Chm70) assumes the fraction initially retained on herbage, F, to be of the form:

$$F = 1 - \exp(-\nu W) \ , \tag{54}$$

where ν is a retention coefficient in m^2kg^{-1} and W is the herbage density in kg dry matter per m^2. Experimental values of ν are 3.3 (Mib65), 2.3 (Chd70), 3.1 (Chm67) and 2.8 m^2kg^{-1} (Chm66a). These results all refer to grassland and there does not seem to be any marked dependence on the on physical form of the deposited activity. (53) is the first term in the series expansion of (54) and a good approximation to (54) when νW is small. Eriksson (Eri77) reports a significant difference between the initial retention of particles on dry and prewetted grass. The initial retention factor increases 2 to 5 times for prewetted grass.

Information from the literature reviewed does not alter the recommendation in WASH-2400 (Was75) of using

$$F = 1 - \exp(-2.8W) \ , \qquad (55)$$

where W is in kg dry matter per m^2.

6.3. Weathering

6.3.1. Introduction.

Many studies have been performed on loss of initially retained material. The weathering mechanisms responsible for removal of material are precipitation and wind. With the effects of radioactive decay included the fraction, F, of the activity initially retained on the vegetation is normally approximately described by an exponential relationship of the form:

$$F = \exp(-\lambda_e t) \ , \qquad (56)$$

where t is the time since deposition and λ_e is a loss-rate constant in units of $time^{-1}$. A frequently used concept is the *weathering halflife*, t_w, which can be determined from the expressions:

$$t_e = \frac{t_w t_r}{t_w + t_r} \quad \text{or} \quad \frac{1}{t_e} = \frac{1}{t_w} + \frac{1}{t_r} \ . \qquad (57)$$

where t_e is the effective residence halflife (also called the residence time) on the vegetation and t_r is the radioactive halflife of the particular radionuclide. It should be checked whether t_e used in the literature is based upon activity per unit area of the ground surface or upon activity per unit weight (dry or wet) of plant matter. In the latter case t_w will be dependent upon dilution of activity by plant growth.

The vertical distribution of contamination in vegetation may be of importance. For example sheep graze deeper than cows. Hence higher concentration in the plant base might be of importance for sheep but not for cows.

6.3.2. Runoff.

In connection with weathering use of the concepts runoff and washoff is not always very stringent.

Runoff should be defined as the excess precipitation which does not remain on the surface upon which the precipitation falls. This means that a runoff situation will reduce the surface concentration of radioactivity. Where runoff was to be incorporated in a consequence model the ground had to be divided into two types: pervious surfaces and impervious surfaces (roads, houses, etc.). For the first type which is to be considered here Ritchie et al. (Rit78) have estimated that it takes rainfall rates above 25 mm/h to produce significant runoff from areas of vegetation. The occurrence of this kind of rainfall in Europe is very infrequent. Therefore, runoff from plants need not be incorporated into a plant transfer model.

"Washoff" denotes the situation where already deposited material is washed off by precipitation. This is considered as a normal part of weathering.

6.3.3. Review of experimental data

Some representative experimental data on retention on vegetation surfaces are presented below.

A series of early experiments are reported by the Agricultural Research Council Radiobiological Laboratory, UK, (Agr60). The results for strontium and caesium are shown in figure 6.1 and give a weathering halflife t_w = 13.9 days on herbage with an average of 11 mm rain per week. Martin (Mat64) reports t_w = 26 days for strontium and 13 days for iodine under arid conditions. Cline et al. (Cli65) report t_w = 8 days for iodine on grass and Heinemann and Vogt (Hei80b) report 7 5 days. For strontium Milbourn and Raylor (Mib65) report t_w = 14 days on the basis of activity per unit area and t_w = 9 days on the basis of activity per unit weight of herbage. They also discovered that as time goes by the strontium content of the basal tissues of the vegetation began to increase and there was a much longer retention

time, more than 100 days, for this part of the vegetation. It is also indicated in the review by Scott Russell (Rus65) that the plant base in general acts as a reservoir with a longer retention time. Kirchmann et al. (Kir66) report t_w ≈ 30 days for strontium and caesium on pastures. Chamberlain (Chm70) derived an average value of 14 days for t_w for pastures during the growing season, and he warns that longer t_w values may be observed in winter. Chadwick and Chamberlain (Chd70) report retention halflives of strontiumchloride of about 19 days in summer and 50 days in winter. Krieger and Burmann (Kri69) made some very interesting experiments on removal of strontium and

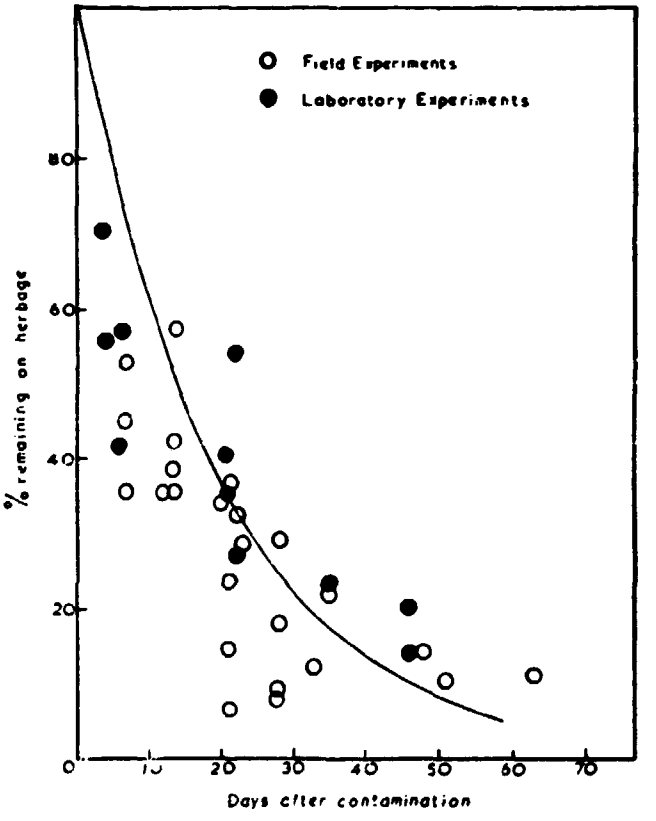

Fig. 6.1. Loss of strontium and caesium from grass. (Agr60).

caesium from pastures. For areas protected against rain one component weathering halflife of 15-18 days was observed, while in the plots exposed to rain the values were 7-10 days for caesium and 3-5 days for strontium. A more thorough statistical analysis showed two effective halflives. The first halflife component for both strontium and caesium averaged 10 and 4 days for protected and open areas respectively, whereas the second component ranged from 25 to more than 50 days for all cases - see figure 6.2.

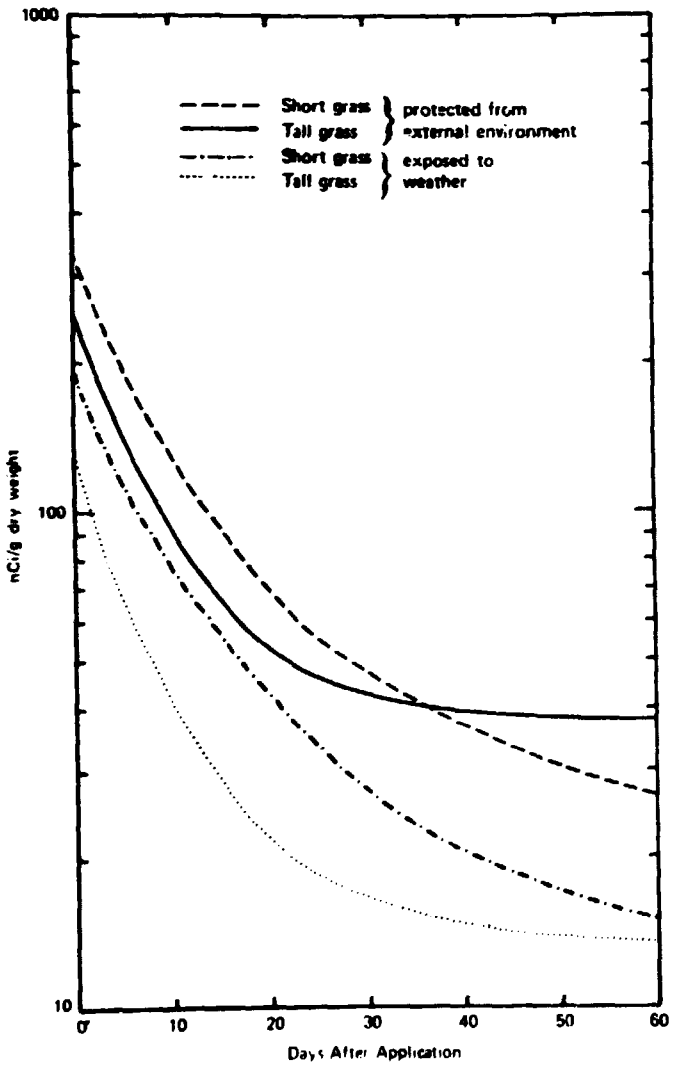

Fig. 6.2. Weathering models derived from experimental data. Soluble ^{85}Sr applied. (Kri69).

Witherspoon and Taylor (Wit70) have made compartment models for their retention data. For their one compartment model they report a weathering halflife of 5-10 days for particle sizes 44-175 μm for different plant species, e.g. soybean and peanuts. Cs-tagged particles, 88-177 μm, were observed (Wit70) to have a weathering halflife of approximately 20 days on meadow grass in the absence of precipitation, and particles were found trapped in the leaf axils. Both Miller (Mie66) and Eriksson (Eri77) report particles deposited during wet or damp conditions as having t_w values twice as great as those for deposition during dry conditions.

6.3.4. Conclusions

For periods longer than one month it is necessary to use a model including at least two weathering halflives and to assume that a certain fraction of the deposited material is not removable by weathering. This fraction is 15% or less (Mie66, Kri69, Was75). For use in a one component exponential function the weathering halflives can be taken from table 5 below.

Table 5. Recommended weathering halflives, t_w.

Situation	t_w [days]	range of [days], measured values
particulates on grass	15	10-27
particulates on vegetation	17	8-28
iodine on grasses	10	7-13

It should also be borne in mind that weathering halflives vary with weather conditions. During extremely dry conditions losses may be appreciable less. Removal by rain will depend not only upon the total rainfall but also upon its intensity and distribution, and the solubility of the material. Also the humidity

(dew) in the periods between showers may affect the retention; there is therefore as expected no simple relationship between meteorological data and rate of loss.

7. RESUSPENSION

7.1. Introduction

Material once deposited on the ground may be resuspended. Considerable attention has been paid to this process, because of the radiological hazards for man from inhalation of radioactive transuranium elements, especially plutonium. Resuspension may obviously also be of importance in the plant transfer model, where the result of these processes will be transfer of material from the ground surface to the external parts of the plants.

This mode of transfer of radioactivity to plants is normally unimportant, but under some circumstances this pathway may be quite significant. In the literature reviewed one case has been found where resuspension is suggested as the major route of contamination of plants. Scott Russell et al. (Rus55) reported this situation as occuring on the Montebello Islands near the west coast of Australia after a nuclear test explosion. The rainfall there is very low, the soil is sandy and dry and the dew is very heavy. When soil and plants were analysed almost the same ratios between different nuclides were found both in soil and plants. This would not have been the situation in the case of root uptake, where some nuclides are absorbed more readily than others. Therefore, it was concluded that the contamination of these plants was mainly due to foilar absorption of material from soil particles blow to the plants.

Resuspension of material from surfaces depends on many factors including the nature of the surface, the age and characteristics of the deposited material and how the material is exposed to

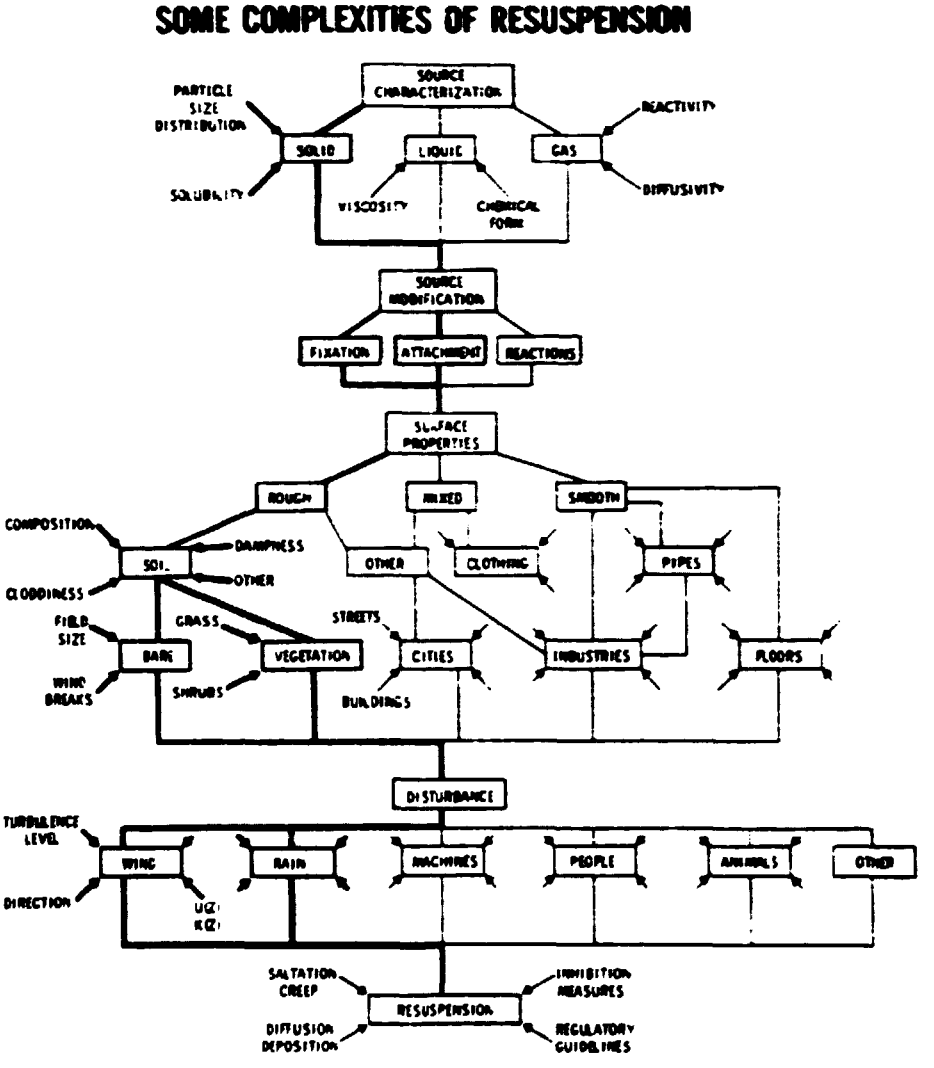

Figure 7.1. Schematic indication of the complexities of resuspension (Sli80).

wind and other physical factors that can initiate the process. Some of the many factors influencing resuspension are shown in figure 7.1 indicating the extreme complexity of the problem.

Resuspension occurs in many places, deserts, rivers, oceans, grasslands, buildings, highways etc.

The overall resuspension processes can be divided into two types, winddriven resuspension, and other mechanical resuspension.

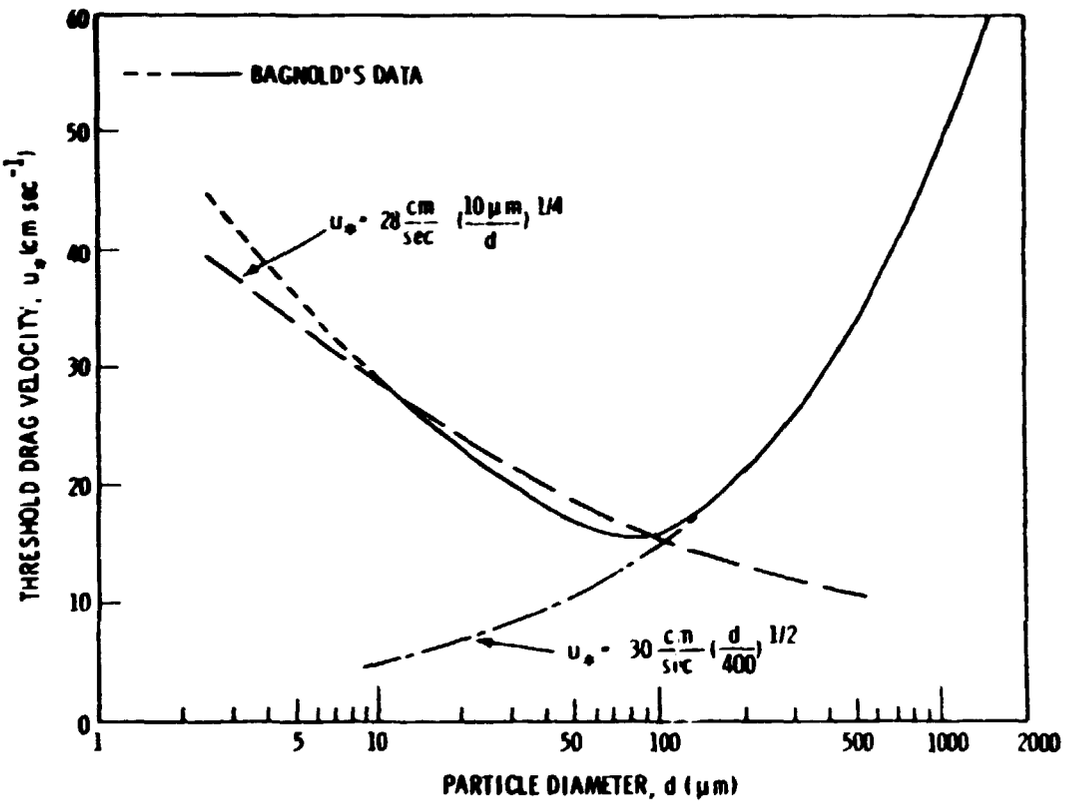

Figure 7.2. Minimum friction velocity required to induce motion of particles, and comparison with theory. (Sli74).

For winddriven resuspension the energy required to dislodge the particles comes from the wind. Figure 7.2 shown the experimentally determined (Bag41) minimum wind stress, expressed by the

friction velocity, u_*, required to induce motion of particles of the indicated size. To move a particle the aerodynamic force must overcome the gravitational force. This explains the behaviour observed for particles greater than 100 μm. For smaller particles the behaviour is explained by Slinn (Sli74). Mechanical resuspension results from mechanical disturbances, e.g. pedestrian or vehicular traffic, digging, ploughing or other agricultural activities.

7.2. Review of experimental data

In this report only winddriven resuspension will be considered. There are no generally accepted models for resuspension. Three different approaches are used for modelling and deriving data on resuspension: the resuspension factor method, the resuspension rate method and the mass loading approach. Each method has its strengths and weaknesses as described below.

The numerous experimental studies can be divided into two different types. The first type uses radioactive tagged particles or other non-active tracers. Only small amounts of material are involved. The second type consists of studies of natural occurring wind blown dust. This latter type has been conducted for a wide variety of surface types and meteorological situations, taking advantage of the fact that the amount of material is seldom a problem. Most experimental data have been reviewed earlier elsewhere (Was75, Lin78a, Las80).

7.2.1. The resuspension factor

Most data on resuspension has been reported in the form of the resuspension factor, K. K is defined as the ratio of the concentration in the air at a reference height (usually one metre) to the concentration of the contaminant per unit surface area and is usually expressed in m^{-1}. The major advantages are its simplicity and the fact that most measurements have been expressed in this form. Disadvantages are that it is a completely empirical formulation, which makes it difficult to extrapolate

Table 6. Experimentally determined resuspension factors. References can be found in WASH-1400 (Was75).

Arid/Desert	Semiarid/Grassland	Urban/Suburban
4×10^{-7} near road graded in fallout field[a]	5×10^{-6} (ZnS) walking and loading boxes[f]	2.5×10^{-6} sandblasting for I-131 removal[g]
3×10^{-5} in Land Rover during travel to work site and outside during work; fission products (5-18 hr)[a]	1×10^{-5} ($\pm0.5\times10^{-5}$) limestone, rock, sand, grass, bushes; fission products[a]	2×10^{-6} 0- to 4-μm U_3O_8 particles on concrete slabs[a]
3 to 10×10^{-4} downwind of crater[a] 1×10^{-3} mechanical disturbance[a] } UO_2	0.8 to 2×10^{-7} crater of tower shot; fission products; road graded from soil disturbance[a]	1.5×10^{-7} 0-to 12-μm U_3O_8 particles on concrete[a]
5×10^{-4} dust from vehicles 10^{-4} to 10^{-6} dust from pedestrians[a] } PuO_2	2×10^{-5} to 4×10^{-6} in vehicle on road graded through fallout[a]	10^{-4} to 10^{-2} fraction resuspended per pass through particles[h]
3×10^{-7} particles of 7 μm MMD on 6-m circles (sandy) furrowed and rocky with sand base (dry)[b]	9×10^{-6} 0 to 4 μm U_3O_8 5×10^{-7} 0 to 12 μm U_3O_8 } undisturbed sandy soil with desert grasses[a]	$K = 2\times10^{-2}$ to 2×10^{-4}; initially very rapid half-time of a few days
1×10^{-7} to 3×10^{-7} (wet)[b]	8×10^{-9} to 10^{-11} Y-91 aqueous solution on sandy soil-rain weathering small plot[a]	$\sim2\times10^{-5}$ to 2×10^{-3} at 4 days
1.2×10^{-7} in 1000-μg/m^2 isopleth 1.4×10^{-7} in 100-μg/m^2 isopleth 1.6×10^{-6} in 10-μg/m^2 isopleth } PuO_2 Safety Test[c]	1×10^{-7} for particles of 7 μm MMD on 6-m circles, grass plot (dry)[b] 4×10^{-8} for particles of 7 μm MMD on 6-m circles, grass plot protected by snow fence (dry)[b]	
~35 day half-time air concentration decay 1 to 3-μm median aerosol size	2×10^{-7} both cases, above, wet[b]	
38-day half-time for resuspension of fission products from Schooner[d]		
76-day half-time for resuspension of fission products from Baneberry[d]		
3×10^{-10} for 8- to 10- year Pu aged deposit[e] 2×10^{-9} for direction of strongest wind[e]		

(a) Stewart (1964).
(b) Healy and Fuguay (1959).
(c) Wilson et al. (1960).
(d) Anspaugh et al. (1973).
(e) Anspaugh (1973).
(f) Schwendiman (1958).
(g) Chamberlain et al. (1951).
(h) Anspaugh et al. (1974).
(i) Fish et al. (1965).
(j) Bailey and Rohr (1953).
(k) Brunskill (1964).
(l) Jones and Pond (1964).

from one terrain to another, and that it ignores the distribution of contamination over the area and the size of the contaminated area involved. The air concentration is dependent on the amount of material that has been resuspended upwind. Resuspension factors for different situations reviewed in WASH-1400 (Was75) are given in table 6. These resuspension factors show large differencies, 11 orders of magnitude, but they include mechanical resuspension and do not specify particle size or density etc.

In the weathering process, the resuspendible fraction of the deposited material changes with time as a result of chemical and physical reactions. Freshly deposited material will have its own characteristics independent of the surface. It will all be available for resuspension. Because of weathering the material becomes more or less attached to the surface, depending upon the type of material and surface. In the time between initial deposition and the development of an aged deposit it has in several cases been observed that the resuspension factor gradually decreases. Kathren and Langham (Kat68, Lah69) originally introduced the time dependent resuspension factor, K(t). This representation is merely of mathematical convenience, since in reality the resuspension factor is unchanged, but the concentration of material available for resuspension decreases. In WASH-1400 (Was75) the following formula for the time dependence of the resuspension factor has been proposed:

$$K(t) = K_e + K_o \exp(-\lambda_t t) \ , \qquad (58)$$

where K_e is the "equilibrium" resuspension factor below which K(t) does not fall and $K_o >> K_e$ is the initial resuspension factor and λ_r is a resuspension decay constant in units of time^{-1}. WASH-1400 suggested $K_e = 10^{-9} m^{-1}$, $K_o = 10^{-5} m^{-1}$ and $\lambda_r = 0.667\ y^{-1}$. Taking $K_o = 10^{-5} m^{-1}$ is more realistic than Anspaugh's (Ans75) recommendation of $K_o = 10^{-4} m^{-1}$, (Gar79, Vas80).

Garland (Gar79) carried out measurements on submicron radioactive tungstic oxide, WO_3, particles in an outdoor wind tunnel on grass and on bare soil. Garland found a resuspension factor between 10^{-7} and $10^{-8}\ m^{-1}$. It increased with the time of exposure to the wind and increased with the equare or cube of the wind speed.

The study by Vashi et al. (Vas80) was carried out to estimate resuspension factors for plutonium from a humid beach silt in the vicinity of a low level liquid effluent discharge point from a fuel processing plant. Their resuspension factors vary

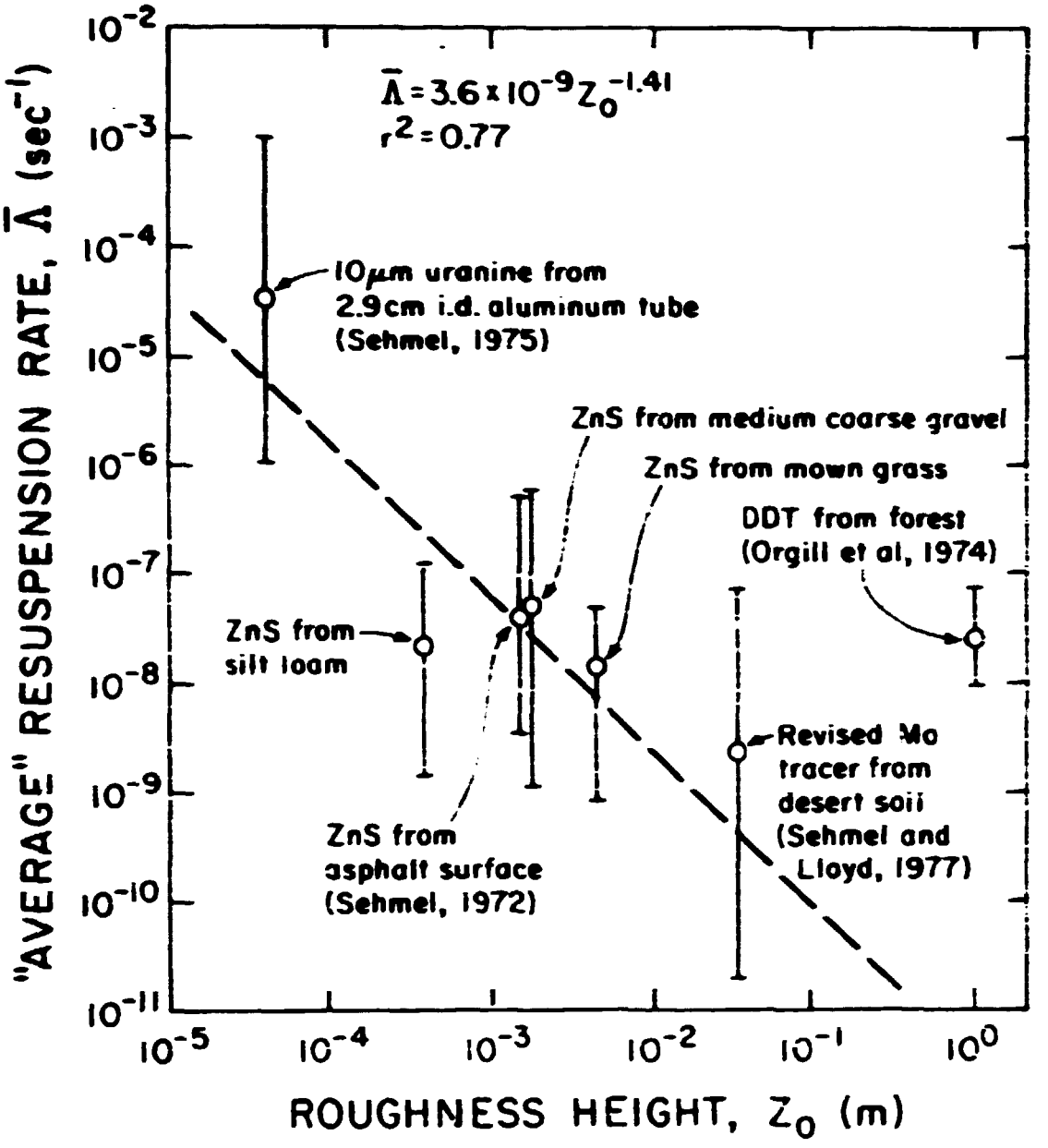

Figure 7.3. Average resuspension rates from various surfaces as a function of surface roughness height. The average resuspension rate and the range of values obtained in each case are indicated. (Sil79).

between 4 10^{-7} and 10^{-8} m^{-1} at wind speeds below 3 m/s. These values are lower than those generally reported. This could be due to the fact, that the resuspended material was deposited in liquid form.

Use of the resuspension factor can be criticised, because the concentration measured at one point is very much influenced by the surface contamination over an unknown distance upwind.

7.2.2. The resuspension rate

An alternative way of expressing resuspension results is in terms of a resuspension rate. The resuspension rate is defined as the fraction of the material present on the ground resuspended per unit time. Experimentally determined resuspension rates have been reviewed by Healy (Hea79). Sehmel (Seh77) observed resuspension rates from 10^{-11} to 10^{-7} s^{-1} at wind speeds up to 6 m/s. The resuspension rates increased with powers of the wind speed varying from 1 to 4.8. The experiments by Garland (Gar79) give a resuspension rate of approximately 2 10^{-9} s^{-1}, which is within the range of Sehmel's observations (Seh77). Slinn and coworkers (Sli79) present a vast amount of experimental results. They resuspended tracer material deposited on a variety of outdoor surfaces using a portable wind generator. For tracer particles, 1-5 μm, average resuspension rates were 1.3 10^{-8}, 2.3 10^{-8} and 5.6 10^{-8} s^{-1} from grass, silt loam soil and gravel, respectively. The dependence on the friction velocity, u_*, was: $R = u_*^{\beta}$, where β goes approximately from 0.5 to 3. Resuspension rates were found to correlate with the roughness height, z_0, of the surfaces, see figure 7.3.

7.2.3. The mass loading approach

Under certain conditions the mass loading approach has been used successfully to predict air concentrations. This concept is an attempt to by-pass the details of the soil characteristics and the resuspension processes. The basic assumption is that the concentration per unit mass contaminant in suspended material is the same as in surface material. A value for the average mass

loading of dust per unit volume of air is assumed or measured. From a measurement of the concentration of contaminant at the surface, the air concentration can then be predicted. This method has been shown to give good agreement with measured values for aged deposits (Ans75). Table 7 shows a comparison of predicted and measured air concentration using an atmospheric mass loading of 100 $\mu g/m^3$, as suggested by Anspaugh (Ans74) for predictive purposes. Predicted values are generally conservative as seen from the results shown.

Table 7. A comparison between predicted and measured air concentrations using a mass loading of 100 $\mu g/m^3$. References can be found in (Hea79).

Reference	Nuclide	Air concentrations		Unit
		Predicted	Measured	
Anspaugh et al. (67)	Pu-239	0.0072	0.0066	pCi/m^3
Anspaugh et al. (67)	Pu-239	0.12	0.023	pCi/m^3
Gudiksen et al. (71)	U-238	150	52	pg/m^3
Gudiksen et al. (72)	U-238	150	100	pg/m^3
Silver et al. (73)	U-238	150	86	pg/m^3
Silver et al. (73)	K-40	0.001	0.00098	pCi/m^3
Sedlet et al. (74)	Th-232	320	240	pg/m^3
Sedlet et al. (74)	U-natural	215	170	pg/m^3
Hamilton (75)	U-natural	110	62	pg/m^3

7.3. Conclusions

The time dependence of the resuspension factor is given by (58). The use of $K_e = 10^{-9}$ m^{-1} and $\lambda_r = 0.677$ y^{-1} in all cases can be recommended. K_o can be set equal to 10^{-4}, 10^{-5} and 10^{-7} m^{-1} for urban areas, bare soil and vegetated surfaces, respectively. It should be emphasized here that the accuracy of any parameterization of resuspension depends on the averaging time. This is also the case in fact for some wet and dry depo-

sition parameterizations, but for resuspension the timescale for the event (e.g. a severe gale) can be close to the averaging time. Suppose that a severe gale lasting one hour resuspends a fraction, f, of the deposited material. This will give a resuspension rate of $f/3600\ s^{-1} \approx f \cdot 3 \cdot 10^{-4}\ s^{-1}$. If this is the only gale during one year the contribution to the annual average resuspension rate becomes $f/31557600\ s^{-1} \approx f \cdot 3 \cdot 10^{-8}\ s^{-1}$. Besides this discrepancy of four orders of magnitude variation there can exist many order of magnitude variations caused by different weather situations, soil types, vegetation types etc. The accuracy of resuspension factors is not better than several orders of magnitude.

8. TRANSLOCATION FROM EXTERNAL TO INTERNAL PARTS OF PLANTS

8.1. Introduction

The contamination on the surface of plants will in general to some extent be removed before consumption by man, for example, by washing or peeling vegetables and fruit, or by removal of the outer protective layers in the production of flour from grain. The amount of material which is transaferred to man will depend on two factors. Firstly it will depend on individual handling and preparation methods. Secondly, it will depend on the amount of material translocated from plant surfaces to the internal tissues of the plants. Material absorbed into the plant is obviously less exposed to loss by the action of wind, rain and food preparation methods. Direct translocation can occur in three ways: through leaves (foilar absorption), through inflorescence (floral absorption), and through the basal parts of the plants or surface roots (plant base absorption). These pathways are distinguished from absorption from the soil via the root system, which has already been discussed in chapter 4. The factors that affect translocation are the characteristics of the deposit (particulate size, solubility, chemical character-

istics etc.), the biological characteristics of the plant (growth habit, physiological status etc.) and the environmental conditions (humidity, temperature etc.). Also the primary mode of deposition is important, since it may greatly affect the availability of the material for absorption by plants. Gaseous iodine is primarily absorbed through stomata. Soluble material is absorbed both through stomata and the epidermis. The absorption of strontium and caesium has been the most extensively studied.

Experimental determination of the amount of material translocated is difficult because there is no unambiguous way of distinguishing between material which has entered the tissue from that which remains on the surface. Different methods for removing the surface material have been used by the different investigators.

8.2. Review of experimental data

In a series of experiments reported by the Agricultural Research Council Radiobiological Laboratory, UK, (Agr60) less than 0.5% of an applied strontium-89 solution was translocated into the primary leaves of wheat plants. Hungate et al. (Hun60) report a 10% translocation of iodine-131 applied during nine days to geranium leaves. Hungate et al. (Hun63) report less absorption of iodine gas at night compared with day, due to night time closure of the stomata. Furthermore they report 5% of the total, absorbed iodine to be translocated to other parts of the plants. Absorbed caesium is reported to be much more mobile within the plant than absorbed strontium (Rus66). Ambler and Menzel (Amb66) present a vast amount of data. They report percentages of foliar-applied Sr-89 as chloride, retained against washing after 3 and 24 hours at different temperatures and different relative humidities for 12 different plant species. The experiments show a wide variation in the translocation rates of strontium into leaves of different plant species. Wettability of the leaf and the relative humidity of the air were important factors affecting the amount absorbed. Increasing the temperature of the air caused some increase in the amount absorbed at the higher relative

humidities, but the temperature had little influence at low relative humidities. Thompson and Howe (Tho73) report 10-25% of deposited iodine-131 on vegetables not to be removable by ordinary household preparation practices. Almost complete translocation of soluble Cs-137 applied to tall fescue grass as particles of 88-177 µm diameter has been reported (Dah69), but this is exceptional and dew has in this case undoubtedly aided the transfer. The amount of caesium translocation is often deduced from the behaviour of potassium and rubidium, elements from the same chemical main group. However, there is evidence that caesium and rubidium behave differently during translocation (Wal70). There is considerable evidence that plant base absorption is of very great importance, particularly with respect to strontium (Rus66). A somewhat special sitation occurs for lichens and mosses. These plants are the first link in what has been proved to be an important food chain in arctic and subarctic regions. They have no root system, and all absorptions take place through the surface. It has been estimated that these plants have 10 times the surface area compared to that of a typical grass. It has been indicated that 95% of the deposited material on lichens and mosses is translocated (Sve65). Also mushrooms have a high translocation of active material and a high Cs-137 burden has been found in those who frequently eat mushrooms (See69). Aarkrog (Aar69, Aar71, Aar75) presents a vast amount of data on the translocation of different radionuclides into rye, barley, wheat and oats. The amount translocated depends on solubility and the time of contamination before harvest.

8.3. Conclusions

It appears that the fraction of deposited material translocated into the internal parts of plants is usually small. It is therefore normally ignored for the majority of elements, except for iodine and caesium. Translocation factors have been given in (Nat79). A maximum translocation factor can be set to 25% and the average value (averaged over materials and plants) lies between 5 and 10%.

9. SUMMARY AND RECOMMENDATIONS

9.1. Introduction

This report presents a plant transfer model. For each transfer pathway in this model theoretical and experimental data are reported. Only a few radionuclides in an effluent release will contribute significantly to internal dose commitments through ingestion. In this study only iodine-131, strontium-90 and caesium-137 have been considered. Below, the data for each transfer pathway are summarized, and recommendations for parameter values for use in dose calculations are given. The recommendations given are as realistic as possible. For specific situations it may often be preferable to choose specific parameter values to obtain the most realistic evaluation of transfer of radioactivity to plants.

9.2. Root uptake (TP4)

Normally, when considering short-term health hazards, transfer of radioactivity from soil to plants via root uptake is ignored without referring to experimental data. On the basis of the literature reviewed it can be confirmed that this transfer pathway is negligible in the short term compared to direct contamination. The maximum root uptake of Cs and Sr for all relevant vegetation during one growing season can be set to 7% of the soil content. No experimental data illustrating in detail the seasonal variation of root uptake have been found in this study.

9.3. Deposition (TP5)

Deposition denotes all processes resulting in direct transfer of matter, gaseous or particulate, from the atmosphere to the ground.

In deposition experiments the experimental variables (wind speed, particle size distribution, vegetation density, surface roughness etc.) are often not very well controlled and/or described. This is also true of experiments on other transfer pathways. The summary and recommendations for deposition are divided into four categories: dry deposition of gases, dry deposition of particles, precipitation scavenging (wet deposition) of gases and precipitation scavenging of particles.

9.3.1. Dry deposition of gases

For dry deposition it is very important to realize that matter cannot be deposited faster than it can be transported to the ground. For gases: such transfer being essentially by turbulent flow, this means that it is always possible to give a maximum deposition velocity, max V_g, on the basis of simple micrometeorological parameters: wind speed, u, and friction velocity, u_*.

The only relevant gas is iodine. Deposition of iodine has been studied extensively. Figure 9.1 gives a summary of iodine deposition velocities (Seh80). The data are shown in order, according to the maximum deposition velocity reported in each experiment. Even though a great amount of experimental material is presented in this review it is difficult to find any consistency in the data. It is difficult to give an average (over all parameters) deposition velocity for elemental iodine, a conservative value is 1 cm/s.

9.3.2. Dry deposition of particles

The variation of dry deposition of particles with particle size is qualitatively well understood. Experimentally determined particle dry deposition velocities range over several orders of magnitude. In figure 9.2 the range of experimental deposition velocities as a function of particle size for each set of experiments is presented (Seh80). Experimental results with polydisperse aerosols are represented by dashed lines. The deposition velocities are lowest for particle diameters in the range 0.1 to

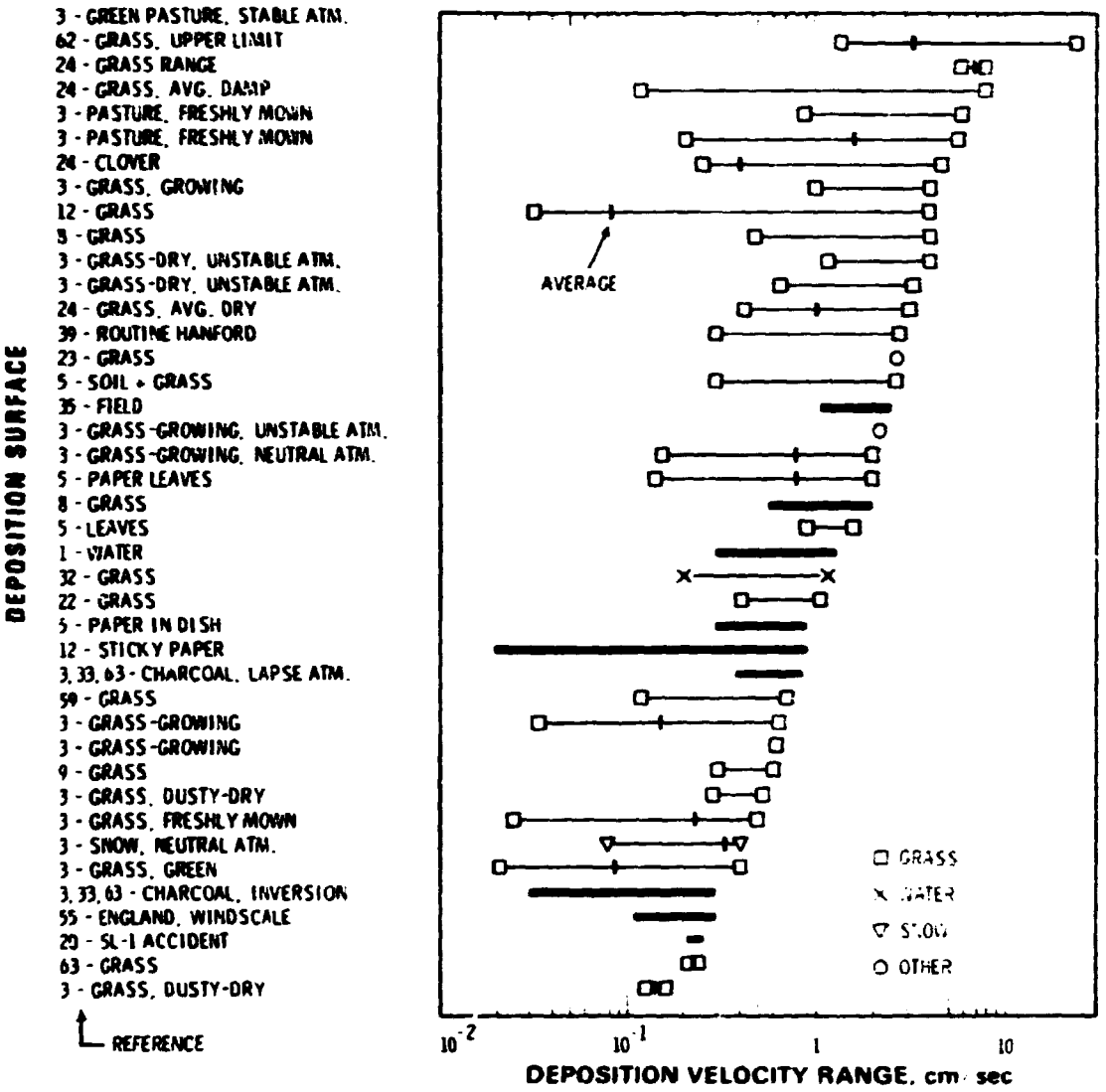

Figure 9.1. Iodine deposition velocities (Seh80) (References in common with this report are: 3=(Bun68), 5=(Chm60), 8=(Chm66a), 9=(Cli65), 22=(Haw64) and 23=(Hei74)).

1 µm. Of course, other parameters also influence these deposition velocities to a great extent. It is difficult to give a recommendation for an average particle deposition velocity. For deposition onto vegetation surface an average V_g value of 1 cm/s can be recommended for particle diameters up to 5 µm. For deposition onto hard surfaces such as roads and roofs V_g will probably be an order of magnitude less. However, experimental data are needed to support this estimate.

9.3.3. Precipitation scavenging of gases

As already mentioned the only relevant gas is iodine. Only very few measurements of iodine scavenging coefficients have been made. Washout coefficient for iodine can be set to 10^{-6} for precipitation intensities up to 5 mm/h. The snowout coefficient for iodine can be set to 10^{-7} for all relevant snowfall rates. These values should be seen as "order-of-magnitude" recommendations and more research in the field is obviously needed.

9.3.4. Precipitation scavenging of particles

The particle and the raindrop size distributions determine the size of the washout coefficients, Λ_p. Λ can be expressed as $\Lambda = c \cdot p^a$, where c is a constant in s^{-1}, p the precipitation rate in mm/h and a is a number between 0 and 1. A typical Λ_p value averaged over relevant particle sizes and for a rainfall rate of 5 mm/h is 10^{-4} s^{-1}. Λ_p will be proportionally smaller for normal precipitation rates of 2 mm/h or less. More than an order-of-magnitude accuracy should not be expected.

In a very recent study (Sli79) in-cloud scavenging is reported to be of the same relative importance as below-cloud scavenging.

According to the literature studied, the use of washout ratios is not to be recommended.

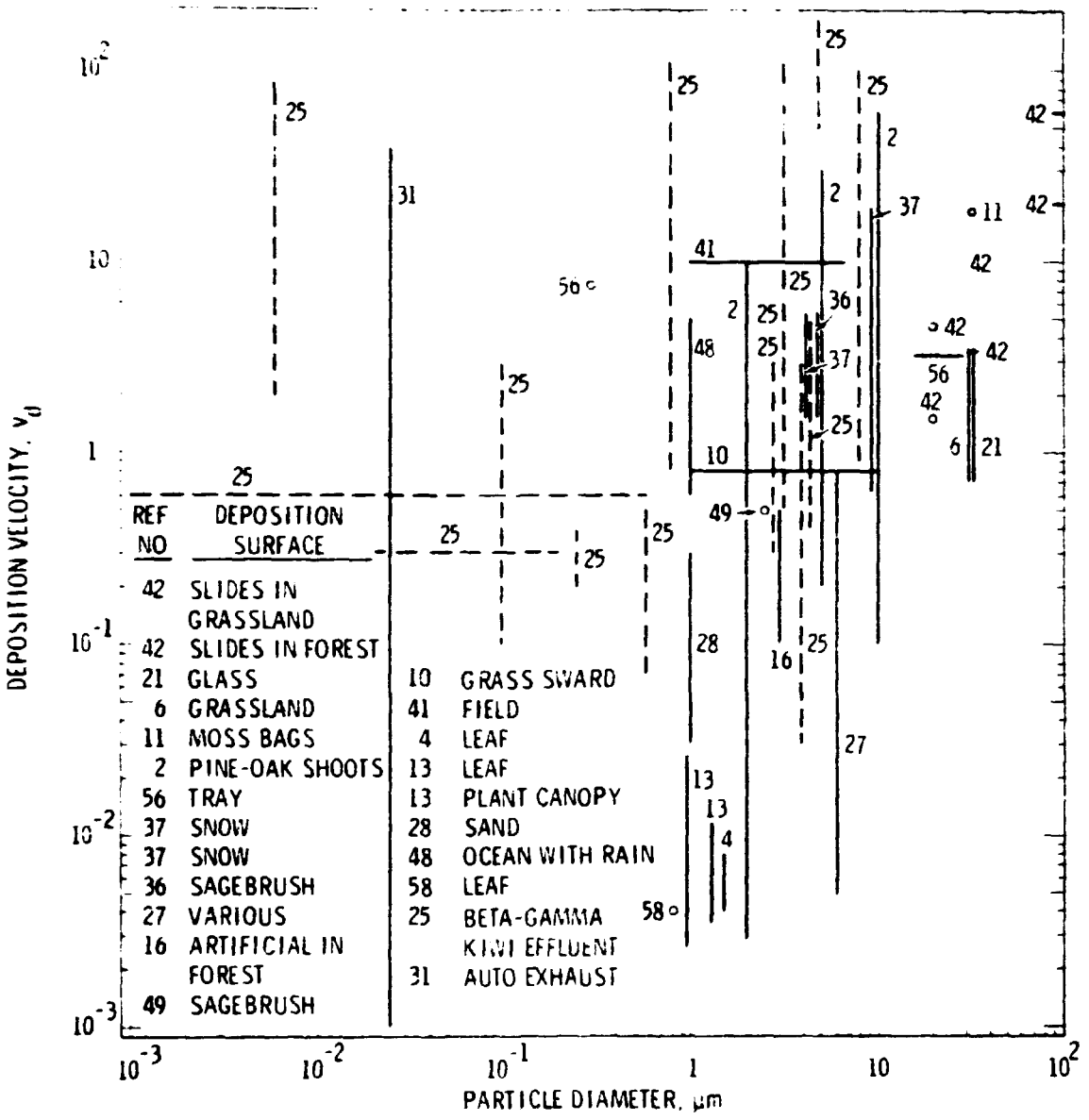

Figure 9.2. Particle deposition velocity summary (Seh80) (References in common with this report are: 6=(Chm67) and 11=(Clo75)).

9.4. Retention on exterior vegetation

Retention of material on vegetation surfaces may be divided according to two time intervals. In the first interval the interception factor is defined as the fraction of the total deposit to a surface initially remaining on the vegetation. Use of interception factors depends on the method by which the used deposition velocities have been derived. The interception factor can be given in the form: $F = 1-\exp(-\nu W)$, where ν is a retention coefficient in m^2kg^{-1} and W is the vegetation density in kg m^{-2}. Typical interception factors lie in the range 0.2-0.3. Dew causes the interception factor to increase by a factor of two.

In the second time interval the *weathering halflife*, t_W, is introduced. For use in a simple exponential function the following weathering halflives can be recommended: t_W = 15 days for particulates on vegetation and t_W = 10 days for iodine on grass. Dew can increase these t_W values by a factor of two.

Runoff is not measured from vegetation surfaces. Runoff will occur on hard surfaces (roads etc.) for rainfall intensities above ~ 3 mm/h. We have not found suffcient data for us to be able to give recommendations for any runoff model.

9.5. Resuspension

Three different approaches are presented for modelling resuspension. Resuspension is time dependent and most data are given in terms of the resuspension factor, K(t). It is here recommended to use $K(t) = K_e + K_o \cdot \exp(-\lambda_r \cdot t)$, with $K_e = 10^{-9}$ m^{-1}, λ_r = 0.667 y^{-1} and K_o equal 10^{-4}, 10^{-5} and 10^{-7} m^{-1} for urban areas, bare soil and vegetated surfaces, respectively.

9.6. Translocation from external to internal parts of plants

Knowledge about the amount of material translocated to the internal tissues of plants is important, since this material will

not be removed by normal handling and preparation methods before consumption by man. It has been found that the fraction of the total deposit on plants, which is translocated into the internal parts of plants is small. Translocation is normally ignored for most elements, except for iodine and caesium. On average (averaged over weather situations, type of material and plant types) the translocation factor will lie between 5 and 10%. Dew will aid translocation.

9.7. Final remarks

The theory of dry deposition mechanisms for all surfaces is well established, whereas a lot more experiments concerning wet deposition on plants and deposition on hard surfaces in general should be done. More experiments on in- versus below-cloud scavenging are needed to estimate the relative importance of these processes. Finally almost no experiments on runoff have been made. Further research on all transfer pathways is needed to give more accurate and reliable recommendations of transfer parameters.

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12. LIST OF SYMBOLS

a	= particle radius, L
a	= constant
α	= constant
B	= Stanton number, dimensionless
B	= biological quality factor in Eqs. (27), (28) and (29)
β	= exponent, dimensionless
c	= constant
C_V	= contaminant concentration on vegetation, M^{-1}
d	= diameter, L
d	= displacement height, L
D	= diffusivity, L^2T^{-1}
E	= collection efficiency, dimensionless
F	= flux (positive down), $L^{-2}T^{-1}$
g	= acceleration of gravity, LT^{-2}
h	= canopy height, L
k	= Kármán constant
K	= resuspension factor, L^{-1}
L	= Monin-Obukhov length, L
λ	= decay constant, T^{-1}
Λ	= scavenging coefficient, T^{-1}
N	= number distribution, dimensionless
ν	= kinematic viscosity, L^2T^{-1}
ν	= retention coefficient in eqs. (53), L^2M^{-1}
p	= precipitation rate, LT^{-1}
r	= transfer resistance, TL^{-1}
Re	= Reynolds number, dimensionless
ρ	= mass density, ML^{-3}
s	= diameter, L
Sc	= Schmidt number, dimensionless
Sh	= Sherwood number, dimensionless
τ	= surface drag, $ML^{-1}T^{-2}$
u	= wind speed, LT^{-1}

u_* = friction velocity, LT^{-1}
μ = retention coefficient, L^2M^{-1}
t_r = relaxation time, T
V = velocity, LT^{-1}
V_i = impact velocity, LT^{-1}
V_D = normalized deposition velocity, $L^3M^{-1}T^{-1}$
V_g = deposition velocity, LT^{-1}
V_s = terminal or sedimentation velocity, LT^{-1}
w = washout ratio, dimensionless
W = grass density, ML^{-2}
W = vertical wind speed in eqs. (40)
W = wet flux, $L^{-2}T^{-1}$
X = contaminant concentration in air, L^{-3}
z_o = roughness length, L
t = halflife, T

Subscripts

d = dry
e = effectiv
e = equilibrium
g = gas
h = hydrometeor
o = ground level
p = particle
p = precipitation
r = rain
r = residence
r = resuspension
s = snow
T = total (chp. 5)
w = water
w = weathering
w = wet

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